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Review

New Trends in Composite Coagulants for Water and Wastewater Treatment

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Abstract: Coagulation/Flocculation (C/F) process aims to efficiently eliminate turbidity, TSS, COD, BOD, toxic metals, phosphates, and UV_{254nm} from wastewater. Both natural and synthetic coagulants, used alone or in conjunction with flocculants, play crucial roles in this treatment. This review summarizes recent trends in coagulants for wastewater treatment, highlighting a wide array of inorganic and organic coagulants that have demonstrated significant efficacy based on reviewed studies. Notably, Crab Shell Bio-Coagulant (CS) excels in turbidity remov5al, achieving a remarkable 98.91% removal rate, while oak leaves protein shows superior performance in TSS and COD removal. Synthetic inorganic coagulants like PALS, $PSiFAC_{1.5:10:15}$, and $PAPEFAC_{1.5\text{-}10\text{-}15}$ demonstrate outstanding turbidity removal rates, over 96%. POFC-2 coagulant stands out for efficiently removing TSS and COD from domestic wastewater, achieving up to 93% removal for TSS and 89% for COD. Moreover, the utilization of $FeCl_3$ as an inorganic coagulant alongside chitosan as an organic flocculant shows promise in reducing turbidity, COD, and polyphenols in wastewater from vegetable oil refineries. PE-2, a novel organic coagulant, demonstrates exceptional efficacy in eliminating turbidity, TSS, COD, and BOD from sugar industry wastewater. Chitosan shows effectiveness in removing TOC and orthophosphates in brewery wastewater. Additionally, CTAB shows high efficiency in removing various toxic metal ions from wastewater. The hybrid coagulants: $PAAP_{0.1,0.5}$ and PPAZF accomplish exceptional turbidity removal rates, approximately 98%.

Keywords: coagulation; flocculation; novel coagulants; natural coagulants; inorganic coagulants; organic coagulants; wastewater

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1. Introduction

In recent times, there has been a growing interest among researchers in addressing environmental concerns associated with the mineral processing industry [1]. A significant area of focus within this realm involves finding efficient methods for treating wastewater generated during mineral processing operations [2].

Researchers have explored various approaches, including physical adsorption [3], coagulation/flocculation [4], membrane separation [5,6], and microbiological methods [7], which have shown promising outcomes in treating wastewater. These methods are favored for their simplicity in operation and the well-established nature of their technologies [8]. Moreover, there has been ongoing exploration into advanced oxidation processes (AOPs) for treating mineral processing wastewater. Examples include photocatalytic processes [9], ozone oxidation [10], and Fenton processes [11].

Coagulation/flocculation is a crucial process in water and wastewater treatment that involves adding a coagulant to a liquid to destabilize suspended particles [12]. This leads to the formation of larger flocs, which adsorb colloidal and dissolved organic matter onto their surfaces and can be more easily removed through sedimentation [13]. This process effectively removes suspended solids, such as biological solids, soil, particles discharged in wastewater, and decaying organic matter, more than 97% [14], and enhances water quality [15] by eliminating impurities such as dyes, up to 100% [16], organic matter [17], bacteria, and viruses by 99% [18]. Additionally, in some instances, coagulation/flocculation can reduce the amount of chemicals needed for treatment, as certain coagulants also act as disinfectants [19]. As a fundamental environmental protection technology, coagulation/flocculation has a wide range of applications in water and wastewater treatment facilities [20]. It is particularly effective for treating surface wastewaters by removing suspended solids (SS) [21], colloidal particles [22], natural organic matter (NOM) [23], biological oxygen demand (BOD) [24], chemical oxygen demand (COD) [25], and other soluble inorganic compounds like phosphate ions [26] and metals like antimony [27]. Research has shown that coagulation/flocculation can also be used as a pre- or post-treatment method for hazardous wastewater. When combined with other treatment processes, such as submerged biological filters (bioreactors), coagulation can significantly enhance treatment efficiency, particularly for refractory wastewaters.

Organic and inorganic materials, used commonly as coagulants and adsorbents, such as aluminum-based [28,29], iron-based [30,31], silica-based [32], are categorized as metal-based coagulants and adsorbents [33]. These coagulants can have detrimental effects, including corrosiveness, high costs, non-degradability, and toxic residues that necessitate special handling [34]. Organic coagulants like polyaluminum chloride (PAC) [35], and polyferric sulfate (PFS) [27] are frequently used because of their high coagulation efficiency at low dosages. However, their use is limited due to their poor biodegradability and low dispersion in water, attributed to their molecular structure [35].

Given the health and environmental issues associated with chemical coagulants, researchers in the water and wastewater industry have been actively exploring non-toxic and environmentally friendly alternatives [36,37]. These alternatives, identified as natural coagulants, are typically classified into plant-origin [38], animal-origin [39], and microorganism-origin [36] types. Natural coagulants are investigated more because of their availableness and properties. They have arisen as a promising way for water and wastewater treatment [40]. These coagulants, chiefly polysaccharides, have established excellent effectiveness in this function. Several advantages are offered, as well as biodegradation, sustainable production, low cost, and they are available [12]. Natural coagulants that are carbohydrates macromolecules, are originating from various plants such as fruits, leaves, seeds, and peels. They entourages physicochemical properties that assist the coagulation flocculation process in water treatment, beyond carboxyl, hydroxyl, and phenolic groups [41].

Hybrid components represent innovative materials with considerable promise for wastewater treatment, offering enhanced efficiency and cost-effectiveness compared to conventional inorganic coagulants and organic flocculants, respectively. The incorporation of functional chemical components into an initial chemical structure to create hybrid materials enhances the aggregation process during coagulation-flocculation in wastewater treatment. Hybrid materials surpass individual components primarily due to the synergistic effects of their elements, which contribute to greater stability compared to standard inorganic coagulants, particularly during storage [42].

The aim of this review paper is to emphasize recently developed natural and chemical coagulants tailored for the treatment of water and wastewater. The review covers the current decade, with an emphasis on the last 3 years. In contrast to earlier studies [43,44], this paper stands out by comprehensively detailing experimental parameters and directly comparing various coagulants, natural, chemical or hybrid, based solely on their removal rates, albeit across different sets of parameters. The diagram in Figure 1 illustrates the different categories of coagulants.

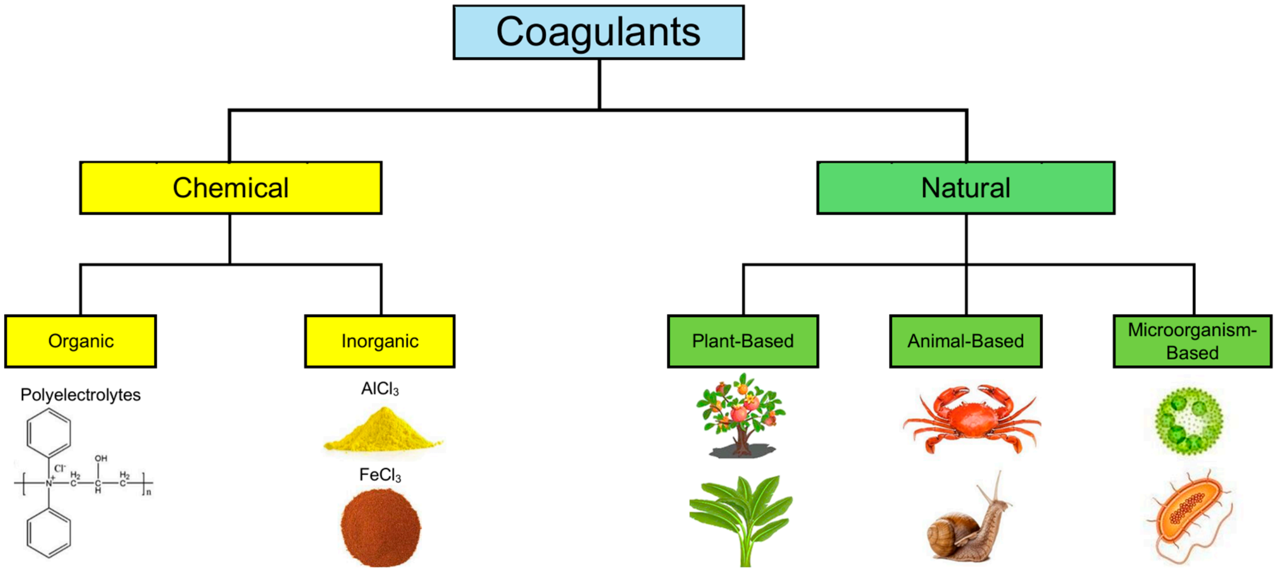

Figure 1. Classification of coagulants and examples.

2. Natural Coagulants

Magnetic rice starch (MS), is a magnetic carbonized starch combining rice starch and Fe_3O_4 in a 1:1 ratio, and was invested by Sibiya et al. [45]. Magnetite (F) was synthesized by mixing 0.4 M Fe^{3+} and 0.2 M Fe^{2+} solutions, agitating, and adding oleic acid, then adjusting the pH to 12 until a black precipitate formed. The precipitate was heated at 70 °C, washed with distilled water and ethanol, and dried at 80 °C for 12 h. Finally, magnetite was combined with rice starch and calcined at 550 °C for 1 h to create magnetic rice starch (MS). The prepared effluent had 11.78 NTU initial turbidity, a TSS concentration of 6.5 mg/L, and COD concentration of 73 mg/L. Coagulation was conducted with stirring at 150 rpm for 2 min (quick mixing), followed by 30 rpm for 15 min (slow mixing). The samples were then allowed to settle for 10 to 60 min, with and without a magnetic field (MF). Results showed that magnetic starch (MS) removed 86% of contaminants (turbidity and TSS) and reduced COD by 55%. Furthermore, MS showed the removal of COD was 56%, turbidity 88%, and TSS 87% when exposed to the magnetic field. The application of magnetic field enhances settling capabilities, resulting in reduced concentrations of suspended particles, and activated sludge, thereby augmenting the efficiency of wastewater management [45]. For several decades, researchers have observed that magnetism is a distinctive property that autonomously contributes to water purification by influencing the physical and chemical characteristics of contaminants in water of contaminants in water. Magnetic exposure to water changes the properties of water and the edge between water and a solid surface sun-drying [46].

Three optimal plants—neem, cassava, and wild betel—were selected as plant-based coagulants (PBC) [47]. Leaves from these locally available plants were collected and dried at 50 °C for 48 h, crushed into powder, and filtered to obtain fine particles. Equal weights of these powdered leaves were mixed thoroughly with distilled water for to ensure homogeneity. The resulting mixture was filtered and used directly for experimentation after preparation to maintain its effectiveness. During the experiment, rapid mixing and slow mixing were conducted at 180 rpm for 3 min and 10 rpm for 20 min, respectively, followed by a 30-min sedimentation period. The properties of the actual wastewater included turbidity ranging from 392.67 to 657.33 NTU, COD concentrations between 711.00 and 727.33 mg/L, TSS levels from 776 to 876 mg/L, and color parameters between 1736 and 1789 ADM at pH 7.13–7.21. The findings showed that plant-based coagulants effectively reduced the measured pollutants, including 85.17% of turbidity, 80.28% of TSS, 53.63% of COD, and 59.42% of color when applied at a dosage of 0.79 mg/L [47].

Crab shells were first washed multiple times to eliminate debris [48]. The cleaned shells were then dried at 220 °C for 15 min and ground into a raw powder (RCS) using a mortar and pestle. To prepare the bio-coagulant (CS), 100 g of RCS was mixed with 1 M sodium hydroxide (NaOH) solution at a solid-liquid ratio of 1:10 (*w*/*v*) in a 1 L beaker. This mixture was stirred for 2 h at 70 °C with a magnetic stirrer. During deproteinization, slight foaming occurred due to the release of proteins from the chitin chains, which eventually subsided. The mixture was cooled to room temperature and allowed to settle for 30 min, then filtered. The filtrate was bleached with a 5% NaClO solution at a solid-liquid ratio of 1:10 (*w*/*v*) for 30 min. The resulting solid could be processed into chitin/chitosan. The experiment conducted with 17.5 mL/L of bio-coagulant, contained 31.6 mg/L of chitosan-protein complex. The optimal removal rates for turbidity were 98.91%, BOD_5 was 92.05%, and COD was 78.92%, were achieved in fish processing wastewater (FPW), >1000 NTU turbidity, 3735 mg O_2/L COD, and 2345.5 mg O_2/L BOD_5 at a pH of 11.3 and a temperature of 25 °C [48].

The *oak leaves* powder coagulant was washed with tap water, dried 24 h to preserve active coagulation compounds, grinded and sieved to achieve a fine, homogeneous powder [49]. This study examines the use of proteins extracted from oak (*Quercus robur*) leaves to treat industrial oily wastewater. The research assesses the removal efficiency for 187 NTU turbidity, 969 mg/L TSS, and 784.45 mg/L COD at a pH of 12 and a coagulant dose of 0.538 mg/L. The results showed removal rates of 96.87% for turbidity, 96.39% for COD, and 89.86% for TSS [49].

The *Moringa peregrina* seed extract was prepared by stirring seed powder in water, with the extraction process conducted at three different temperatures (20 °C, 40 °C, and 60 °C) before filtering the extract [50]. Each mL of this solution corresponded to 10 mg of dry seeds. The *Moringa* seed powder was extracted with 100 mL of 0.1, 0.5, and 1 M NaCl solutions to identify the optimal extraction conditions regarding temperature and salt concentration. To remove the oil, crushed seeds were mixed with petroleum ether and stirred. The oil-extracted seeds were then filtered and left to dry overnight at room temperature. The dried seeds were then extracted with extracting solution under the optimal conditions (60 °C and 0.5 M NaCl) for 1 h. Each mL of this solution equaled 10 mg of dry Moringa seeds. The resulting raw crushed seed extract after oil removal (MPDOEx) were used as coagulants in the coagulation experiment. The initial measurements were 99.1 NTU for turbidity and 188 mg/L for COD. At an optimal seed dosage of 400 mg/L and a pH of 9, the removal efficiency were 38% COD, and 81% of turbidity. The initial concentrations of the metals were as follows: 2.1 mg/L for cadmium, 3.1 mg/L for cobalt, 11.0 mg/L for chromium, 8.1 mg/L for copper, 10.1 mg/L for manganese, 71.8 mg/L for molybdenum, and 3.8 mg/L for nickel. Manganese and nickel exhibited minimal removal efficiency, both being below 15%. However, the other metals showed moderate to high removal rates. The de-oiled Moringa peregrina seed extract, at a dose equivalent to 200 mg/L of raw seeds, was able to remove 97.4% of molybdenum, 66.5% of copper, 51.8% of cadmium, 50.3% of chromium, and 45.8% of cobalt [50].

Prickly pear (PP) fruit peel mucilage gel was assessed as a new coagulant for wastewater treatment [51]. Hydrated prickly pear fruit peels were manually squeezed to obtain a highly viscous gel. To this gel, 95% ethanol was added, resulting in the formation of a milky white supernatant, which corresponded to the mucilage. The mucilage was dried, then reconstituted in distilled water to form a gel-like substance, which was used as a coagulant. The domestic wastewater collected at the tertiary treatment inlet showed high turbidity, 88.00 NTU, and colour, 671 TCU. Coagulation experiments using a coagulant dose of 12 mg/L at a pH of 13 achieved 94% turbidity removal and 85% color removal efficiency [51].

Almond (*Prunus dulcis*) hull and also cherry (*Prunus avium*) pit were preferred as plant-based coagulants [52]. The almond hulls and cherry pits were soaked and dried in an fryer at 70 °C for 1 day. Each natural coagulant was then edged into a powder, cool naturally, and kept. The cattle wastewater had parameters of 7207 NTU turbidity, 21,178 mg O_2/L COD, and 6930 mg/L TSS. The experiments tested in pH 3, 6, 7, and 9. The results demonstrated that using 0.1 g/L This efficiency can be enhanced to 58.2l at pH 3, it was possible to achieve removal rates of 39.1% for COD, 38.3% for turbidity, and 52.9% for TSS. In contrast, using 0.1 g/L cherry pit at pH value of 3, the removal efficiencies were 42.4% for COD, 88.8% for turbidity, and 22.3% for TSS [52].

Moringa oleifera seeds powder (MOSP) served as the coagulant to remove 7.88 mg/L Amido Black 10B dye and high level of turbidity [53]. To eliminate any remaining moisture of these seeds, they were extracted from the pods and subjected to drying in an oven at 50 °C for one day. Before grinding, the seed shells were removed. The results revealed that both turbidity and dye could be completely removed under specific conditions: 0.34 mg/L MOSP dosage, 7.88 mg/L dye concentration at pH 6.93. Experimental findings showed that the removal of turbidity was 98.5% in synthetic wastewater and the dye removal was 92.2% [53].

The prevalent *Avicennia marina* plant, belonging to the *Verbenaceae* family, dominates 97% of the total mangrove habitats in the state [54]. The shed leaves of *A. marina* were washed, dried, and subsequently shredded and ground into powder. Subsequently, the AMC underwent chemical treatment individually by the addition of 0.05 M HCl, 0.05 M NaOH, and 0.5 M NaCl. The mixtures of AMC and hydrolyzing agents were agitated. The filtrate from these mixtures was collected, and the remaining residues were labeled as HCl-treated AMC, NaOH-treated AMC, and NaCl-treated AMC, respectively. For the mud water, the initial turbidity was measured at 15.15 NTU, with an ideal pH of 7.82. In contrast, for the starch water, the initial turbidity was slightly higher at 16.36 NTU, with an ideal pH of 7.9. The dosage of all AMCs tested was maintained at 1.0 g/L. In mud water, the coagulant activities of various AMCs were observed to range from 89.74% to 95.87%, with native AMC showing the lowest activity and HCl treated AMC exhibiting the highest. Similarly, in starch water, coagulant activities ranged from 89.88% to 96.90%, with HCl treated AMC demonstrating the highest activity [54].

The study of Ovuoraye et al. 2021 [55] investigates the effectiveness of Eggshell Coagulant (ESC) in the coagulation-flocculation treatment of cosmetic wastewater. The process of preparing the coagulant from Eggshell involved sun-drying, also used for industrial drying processes [56], for approximately 3 weeks. After subsequent drying, the samples were crushed and ground to improve their surface area. Afterward, they underwent screening to form a finer particle flour. The examination of the wastewater sample reveals a total suspended solids (TSS) concentration of 232 mg/L. The ideal pH and dosage are determined to be 6 and 0.2 g/L, respectively. At this optimal point, there is an 85% removal efficiency for TSS, resulting in a residual TSS concentration of 38 mg/L after treatment with ESC [55].

Aloe vera, harvested from Algerian fields, was prepared for use a coagulant; washed with tap water, dried to preserve active components, grinded, and sieved [57]. Subsequently, *Aloe vera* powder was combined with water and stirred to extract the active coagulants. Once settled, the supernatant was passed through a filter to gain the liquid coagulant. The coagulation process performed well at pH 7 with an initial turbidity of 13 NTU. Optimal dosages of 10 mg/L of *Aloe vera* powder (AV-Powder) and 0.1 mL/L of liquid *Aloe vera* (AV-H_2O) were used. Results indicated that employing the natural coagulant. AV-Powder reduced water turbidity by 28.23% and AV-H_2O by 87.84% [57].

Dillenia indica fruits were collected and rinsed with distilled water [58]. The seeds were manually separated, air-dried for 15 days, ground into powder, and sieved. For extraction, seed powder was oil extracted using ethanol over three cycles until the ethanol became colorless. The seed residue was dried at room temperature. To prepare the crude coagulant, residue was dissolved in distilled water, mixed, and filtered through muslin cloth. The

D. indica seeds surface area was 1.6735 m^2/g and the pore volume was 0.0022 cm^3/g. The average concentrations of Bisphenol A and DEHP in the leachate were 31 mg/L and 15 mg/L, respectively. Under optimal conditions, pH 8.5 removal efficiencies of 60% for 4,4′- (1-methylethylidene)bis- (Bisphenol A) with dosage of 1066 mg/L, and 55% for bis(2-ethylhexyl) phthalate (DEHP) with a dosage of 958 mg/L were achieved [58].

Cassava peel starch (CPS) shows potential as an alternative to alum coagulant for removing turbidity, TSS, and COD from levels 194 NTU, 284/L, and 296 mg/L, respectively [59] Raw CPS samples were sourced from a small-medium industry and processed to eliminate impurities. The procedure involved screening, filtration, peeling, and drying the cassava peels. First the peels were washed with tap water, second rinsed with distilled water and also filtered and dried to eliminate any remaining moisture. The dried peels were blended into a starch slurry, which was then filtered and placed in a settling tank. The starch residue was isolated from the slurry and underwent an additional wash with distilled water. Subsequently, the starch residue was dried, while the peels were finely ground into powder. An alum solution was created by dissolving aluminum sulfate hydrate in distilled water. Similarly, a stock solution of CPS coagulant was made by dissolving CPS powder in distilled water, with vigorous mixing. Doses for each test were determined from the stock solution, with mixtures ranging from 0% to 100% CPS to alum ratio. The best removal of turbidity, total suspended solids (TSS), and chemical oxygen demand (COD) occurred at pH 6.0, achieving removal rates of 60.19%, 57.79%, and 30.19%, respectively, with a CPS concentration of 448.58 mg/L. At pH 8, with a 4:1 ratio of CPS to alum, the removal efficiencies increased to 77.48% for turbidity, 77.34% for TSS, and 56.89% for COD [59].

Table 1 compares all the data of the novel coagulants and the Figure 2 compares the removal rate of turbidity, TSS, and COD. All of the natural coagulants remove efficiency the turbidity, but the most effective is Crab Shell Bio-Coagulant (CS), reach 98.91% turbidity removal [48]. Furthermore, *oak leaves* protein was removing the highest TSS and COD concentration [49]. In all of this research, the wastewater that used to be removed, were rice starch-containing effluent [45], fish processing [48], industrial oil [49], domestic [51], cattle [52], synthetic for dye removal [53], mud and starch water [54], cosmetic [55], leachate contained bisphenol A and DEHP [58].

Table 1. Novel natural coagulants for wastewater treatment.

Coagulants	Remove	Initial Conditions	pH	Coagulant Dosage	Removal	Ref.
MS [1] MS [1] with magnetic exposure	Rice Starch-Containing Effluent	11.78 NTU turbidity 73 mg/L COD 6.5 mg/L TSS	7.2	300 mg/L	86.00% turbidity 85.00% TSS 55.00% COD 88.00% turbidity 87.00% TSS 56.00% COD	[45]
PBC [2]	General Wastewater	392.67–657.33 NTU turbidity 711.00–727.33 mg/L COD 776–876 mg/L TSS 1736–1789 ADM Color	7.13–7.21	0.79 mg/L	85.17% turbidity 54.63% COD 80.28% TSS 59.42% Color	[47]
CS [3] bio-coagulant	Fish Processing Wastewater (FPW)	>1000 NTU Turbidity 3735 mg O_2/L COD 2345.5 mg O_2/L BOD_5	11.3	31.6 mL/L	98.91% turbidity 78.92% COD 92.05% BOD_5	[48]
oak leaves protein	Industrial Oily Wastewater	187 NTU Turbidity 969 mg/L COD 784.45 mg/L TSS	12	0.538 mg/L	96.87% turbidity 96.39% COD 89.86% TSS	[49]
MPDOEx [4]	Synthetic water	99.1 NTU Turbidity 188 mg/L COD 71.8 mg/L Mo 8.1 mg/L Cu 2.1 mg/L Cd 11.0 mg/L Cr 3.1 mg/L Co 10.1 mg/L Mn 3.8 mg/L Ni	9	400 mg/L 200 mg/L	81.00% Turbidity 38.00% COD 97.40% Mo 66.50% Cu 51.80% Cd 50.30% Cr 45.80% Co 12.00% Mn 10.50% Ni	[50]
PP [5] fruit peel mucilage gel	Domestic Wastewater	88 NTU Turbidity 671 TCU color	13	12 mg/L	94.00% turbidity 85.00% color	[51]

Table 1. *Cont.*

Coagulants	Remove	Initial Conditions	pH	Coagulant Dosage	Removal	Ref.
Almond hull (*Prunus dulcis*) cherry pit (*Prunus avium*)	Cattle Wastewater	7207 NTU turbidity 21178 mg O_2/L COD 6930 mg/L TSS	3	0.1 g/L	38.30% turbidity 39.10% COD 52.90% TSS 88.80% turbidity 42.40% COD 22.30% TSS	[52]
MOSP [6]	Synthetic Wastewater	Turbidity 7.88 mg/L Amido Black 10B dye	6.93	0.34 mg/L	98.50% turbidity 92.20% Amido Black 10B	[53]
AMC [7] HCl treated AMC [7] NaOH treated AMC [7] NaCl treated AMC [7]	Starch wastewater Mud Wastewater Starch wastewater Mud Wastewater Starch wastewater Mud Wastewater Starch wastewater Mud Wastewater	16.36 NTU turbidity 15.15 NTU turbidity 16.36 NTU turbidity 15.15 NTU turbidity 16.36 NTU turbidity 15.15 NTU turbidity 16.36 NTU turbidity 15.15 NTU turbidity	7.90 7.82 7.90 7.82 7.90 7.82 7.90 7.82	1.0 g/L	89.88% turbidity 89.74% turbidity 96.77% turbidity 95.87% turbidity 96.90% turbidity 94.73% turbidity 89.87% turbidity 94.66% turbidity	[54]
EC [8]	-	232 mg/L TSS	6	0.2 g/L	85.00% TSS	[55]
AV [9]-powder AV [9]-H_2O	General Wastewater	13 NTU turbidity	6 7.5	10 mg/L 0.1 mL/L	28.23% turbidity 87.84% turbidity	[57]
Dillenia indica	Leachate	31 mg/L Bisphenol A 15 mg/L DEHP	8.5	1066 mg/L 958 mg/L	60.00% Bisphenol A 55.00% DEHP	[58]
CPS [10] CPS [10]:alum (4:1)	Water with Initial Turbidity, TSS, and COD	194 NTU turbidity 284 mg/L TSS 296.248 mg/L COD	6.0 8.0	448.58 mg/L 80% of CPS 20% of alum	60.19% turbidity 57.79% TSS 30.19% COD 77.48% turbidity 77.34% TSS 56.89% COD	[59]

[1] MS: Magnetic rice starch, [2] PBC: plant-based coagulants, [3] CS: Crab shells, [4] MPDOEx: *Moringa peregrina* raw crushed seed extract after oil removal, [5] PP: Prickly pear, [6] MOSP: *Moringa oleifera* seeds powder, [7] AMC, [8] ESC: Eggshell Coagulant, [9] AV: *Aloe vera*, [10] CPS: Cassava peel starch.

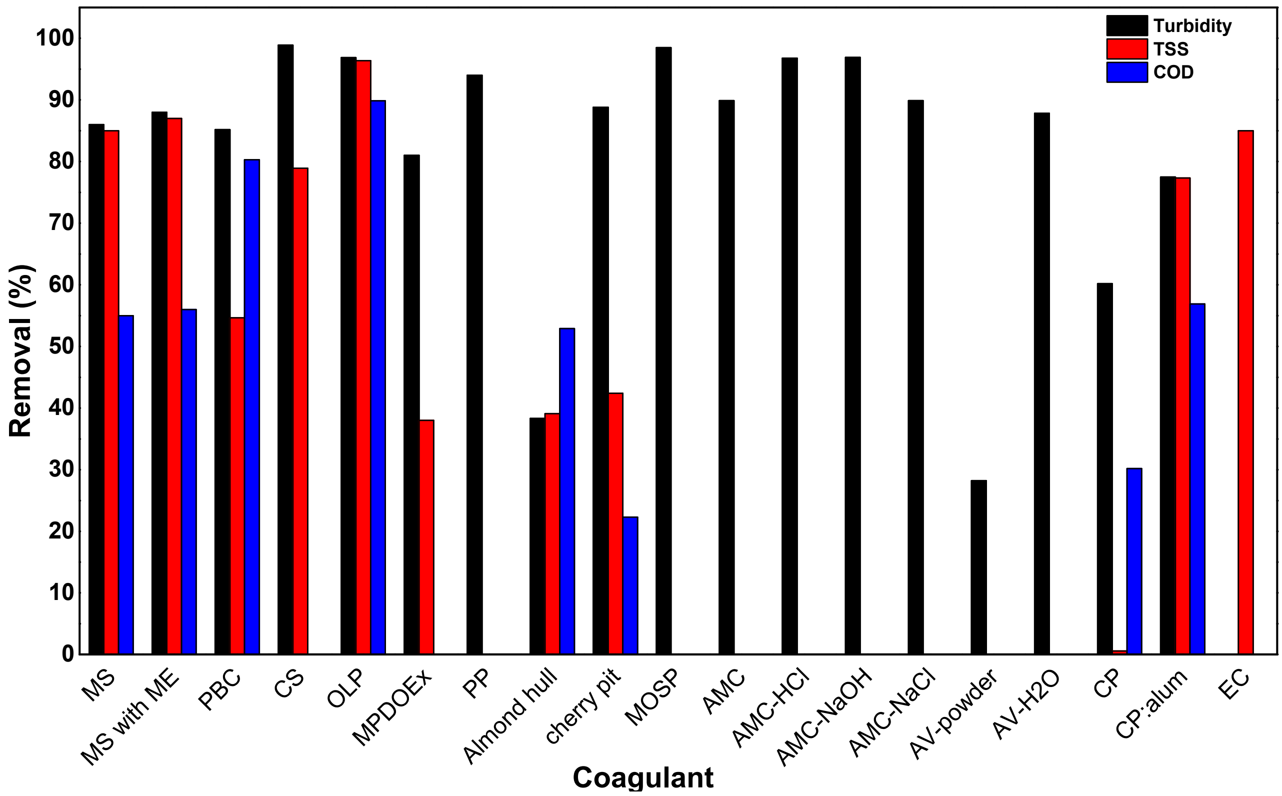

Figure 2. Comparison of novel natural coagulants as per removal rate for turbidity, TSS, and COD.

Natural Coagulants with Flocculants Added

The coagulant was derived from the mucilage content found in prickly pear peel waste [60]. The peel waste underwent washing and cleaning with tap water followed by deionized water. Next, the waste was blended with the deionized water using a blender and heated at 50 °C for an hour. After cooling to room temperature, absolute ethanol was then added at a 1:3 (*v*/*v*) ratio, and the mixture was left to settle for 24 h at refrigerator. Following this, the mixture underwent vacuum filtration to obtain the mucilage extract, which was subsequently dried at room temperature for 24 h. Finally, the dried extract was ground into a powder using a mortar and stored in the refrigerator. The production yield of the coagulant was estimated to be 0.25% *w*/*w*. In these experiments, added 1 mg/L flocculant, a commercial anionic polymer The findings indicate that the most effective conditions for turbidity removal, achieving 76.1%, occur at a water pH of 4 with a coagulant dosage of 100 mg/L. However, when the wastewater pH is adjusted to 7.8 and the coagulant dose is increased to 250 mg/L, the efficiency decreases to 51.7%. This efficiency can be enhanced to 58.2% by employing a blend consisting of 30% coagulant and 70% aluminum sulfate at optimal proportions [60]. According to Aguilera Flores et al., 2024, bio- coagulants are investigated within dual systems alongside chemical coagulants like aluminum salts to enhance their efficiency in reducing water turbidity, while also minimizing the quantity of chemicals of agents used and of the production of sludge.

Table 2 depicts the two different natural coagulants, used with an anionic polymer as flocculant to remove turbidity. For the Prickly pear peel waste, the best removal was at pH 4 with the removal rate being 76.1% [60]. Otherwise, the coagulant that derived from the mixing of 30% prickly pear peel waste and 70% aluminum sulfate achieved 58.1% removal of turbidity at pH 7.8 [60].

Table 2. Novel natural coagulants in combination with flocculants for wastewater treatment.

Coagulant	Flocculant	Initial Conditions	pH	Coagulant Dosage (mg/L)	Flocculant Dosage (mg/L)	Removal	Ref.
Prickly pear peel waste	Anionic polymer	45.39 NTU Turbidity	4	100	1	76.10% turbidity	[60]
			7.8	250		51.70% turbidity	
30% prickly pear peel waste and 70% aluminum sulfate						58.10% turbidity	

3. Inorganic Coagulants

Ali et al. [19] investigated an innovative polymeric inorganic coagulant-flocculant called poly-ferric chloride (POFC), which was prepared using iron-containing waste [19]. Blast furnace iron or pickling waste, along with industrial-grade sodium carbonate from Solvay Alexandria Trading L.L.C., Egypt, are utilized as low-cost raw materials to produce various polymeric coagulants. These coagulants have the general structure $Fe_n(Cl_x(OH)_{3-x})_n$ [19]. In this study evaluated the efficiency of POFC-1 and POFC-2 coagulants in removing turbidity, TSS, and COD from domestic wastewater. The difference between the two coagulants is the ratio of OH/Fe. Coagulation, flocculation, and sedimentation experiments were performed using 10 mg/L (0.1 mL/L) of various POFCs without any added flocculant. The experiment conducted with a rapid mixing speed of 300 rpm for 4 min followed by a sedimentation time of 30 min The influent wastewater had a COD of 342 mg/L, TSS of 300 mg/L, and turbidity of 86 NTU. The results showed that POFC coagulants performed highly effectively in treating domestic wastewater, achieving removal rates of 82–93% for TSS, 78–92% for turbidity, and 67–89% for COD. It was observed that increasing the OH/Fe ratio (basicity) from 0.8 to 2 decreased the efficiency due to reduced charge density on the POFC surface and diminished neutralizing capability. No further improvement was noted with higher POFC basicity. POFC-1-0.8 and POFC-20.8 exhibited superior performance in removing TSS, turbidity, and COD compared to other formulations, with only slight differences in their removal efficiencies [19].

The study of examined the combined removal of Sb(III) and Sb(V) from wastewater using polymeric ferric sulfate (PFS) and an oxidation process [27]. The findings revealed that PFS was more effective at removing Sb(III) than Sb(V) in terms of coagulant dosage, pH conditions, and resistance to interference. In a pure water matrix with 1 mg/L of Sb, a PFS dose of 60 mg/L at a final pH of 5.0 resulted in 95% removal of Sb(III) and 90% removal of Sb(V). Additionally, experiments conducted with printing and dyeing wastewater (PDW) containing 0.085 mg/L Sb(III) and 0.1 mg/L Sb(V) showed that pre-reducing Sb(V) to Sb(III) significantly enhanced the total Sb removal, increasing it from 72.0% to 97.5% During the experiment, the presence of Sb(V) was rapidly detected upon the addition of PFS, indicating the partial oxidation of Sb(III) during the coagulation. The oxidation dynamics of Sb(III) and the fluctuation in Sb(V) concentration were influenced by both the dosage of PFS and the pH conditions. For instance, when 10 mg/L PFS was used, the concentration of Sb(V) peaked at 36.5 μg/L after 5 min, minimizing to 19.0 μg/L after 1 h. Raising the PFS dosage to 60 mg/L led to an elevated peak concentration of Sb(V) reaching 36.5 μg/L within 10 min, followed by a subsequent decline to 13.1 μg/L. The hydrolysis products of PFS functioned as catalysts in the oxidation of Sb(III), a mechanism facilitated by either dissolved oxygen or abiotic electron transfer from Sb(III) to Fe(III). The pH levels, particularly in the range of 9–11, were found to promote higher concentrations of Sb(V), with levels peaking at 81 μg/L at 10 min [27].

Composite coagulants $FeCl_3$–$AlCl_3$ and $FeSO_4$–$Al_2(SO_4)_3$ were prepared by mixing solutions of ferric chloride ($FeCl_3$) with aluminum chloride ($AlCl_3$), and ferrous sulfate ($FeSO_4$) with aluminum sulfate ($Al_2(SO_4)_3$) [61]. The mixture was quickly stirred for 1 min at 200 rpm, then slowly stirred for 30 min at 30 rpm, followed by a 30-min settling period. 0.519 mg/L of total phosphorus (P) was removed using two coagulants, $FeCl_3$–$AlCl_3$ and $FeSO_4$–$Al_2(SO_4)_3$. At pH levels of 5 and 7, these coagulants eliminated 91.31% and 86.82% of the total phosphorus, respectively [61].

The polyaluminum silicate sulfate (PASS) utilized to remove 196.67 mg/L of COD_{Cr} from coking wastewater [62]. In was synthesized PASS in the laboratory following a modified procedure, resulting in an Al/Si ratio of 5. A concentration of 7 mmol/L PASS and pH 7, and resulted in reducing the COD_{Cr} level from 196.67 mg/L to 59.94 mg/L, corresponding to a reduction of 69.5% [62].

Waste aluminum foil was utilized as the primary source of aluminum to produce PACl coagulant [63]. Initially, the granular aluminum foil waste underwent a cleaning process with hot water to eliminate pollutants, followed by drying at 60 °C. The drained aluminum foil underwent hydrolyzation with hot HCl solution while continuously stirring, resulting in the formation of aluminum chloride solution. Subsequently, the reaction temperature of the aluminum chloride solution was raised to 75 °C with stirring. Dropwise addition of NaOH solution under continuously stirring raised the pH to 9 and attained a 2:1 molar ratio of OH to Al, facilitating the format of polyaluminum chloride (PACl) coagulant in the form of a white gel product. After aging for 24 h, the gel was separated through centrifugation, washed multiple times with deionized water, and after dried at 85 °C in air. Under the ideal parameters, which include a pH of 6.5, a temperature of 35 °C, and specific mixing conditions involving rapid agitation for 2 min at 280 rpm followed by slower mixing for 30 min at 80 rpm, along with a settling period of 30 min thereafter, over 98% of turbidity and approximately 69.8% of DOC impurities can be effectively eliminated using a reduced dosage of 25 mg/L PACl coagulant [63].

In accordance with He et al. 2023 [64], Polyaluminum Lanthanum Silicate Coagulant, PALS, was synthesized via co-polymerization to act as a novel coagulant. To prepare the coagulants, pH of a 2.0% sodium silicate solution (*w*/*w* Si) was adjusted to 3. This solution was then stirred vigorously and left to stand. Aluminum chloride ($AlCl_3$) and lanthanum chloride ($LaCl_3{\cdot}7H_2O$) were added to the solution with continuous stirring. After 1 h of stirring, sodiumbicarbonate ($NaHCO_3$) was gradually introduced. The mixture was then stirred for another 3 h and aged for 12 h. The resulting products were dried in a vacuum dryer. Those dried composites were then ground and screened, resulting in PALS. In the coagulation experiments, simulated water was prepared using kaolin, humic acid, and potassium phosphate monobasic (KH_2PO_4). Kaolin was used to mimic turbidity, humid acid to present organic matter, and KH_2PO_4 to pretend total phosphorus (TP). The parameters of simulated wastewater were 28.6–30.2 NTU turbidity, 2 mg/L TP, and 29.68–30.42 mg/L dissolved organic carbon (DOC). The maximum turbidity reduction, approximately 99%, was achieved with a dosage of 12 mg/L at a pH of 8. At pH 8, the removal efficiency of dissolved organic carbon (DOC) was only around 52% with a dosage of 14 mg/L, whereas it significantly increased to approximately 70% at pH 3–7. These findings indicate that acidic conditions are more favorable for the removal of macromolecular organics and colloids, while alkaline conditions are less conducive to dissolved organic matter removal. At a dosage of 8 mg/L and pH 6, the maximum removal efficiency for total phosphorus (TP) could achieve nearly 99.6% [64].

To prepare the poly-ferric-titanium-silicate-chloride (PFTSC) and poly-titanium-silicate-chloride (PTSC) coagulants, an aqueous solution of sodium metasilicate nonahydrate Na_2SiO_3 was first added dropwise to an aqueous HCl solution until the pH reached 4, followed by continuous stirring to activate the polysilicic acid (PSA) solution. Separately, NaOH solution was adjusted to a titanium tetrachloride ($TiCl_4$) solution or a combination of $TiCl_4$ and iron trichloride ($FeCl_3$) while stirring continuously to adjust the alkalinity, resulting in the formation of polytitanium-chloride (PTC) and poly-ferric-titanium-chloride (PFTC) solutions. Subsequently, a measured amount of either PFTC or PTC solution was added to the activated PSA solution. The resulting mixture was stirred and allowed to mature at room temperature to produce the PFTSC and PTSC coagulants. The wastewater had a pH of 9, with initial turbidity of 40 NTU, DOC concentration of 480 mg/L, and COD level of 2000 mg/L. When the dosages of both coagulants were 800 mg/L, PFTC achieved a turbidity removal rate of 93.2%, COD removal rate of 10.7%, and DOC removal rate of 10.1%. Meanwhile, PTSC showed a turbidity removal rate of 80%, with slightly lower removal rates for organic matter compared to PFTC [65].

The targeted coagulants, featuring a long and flexible chain of polyacrylic acid (PAA), contain many –COOH groups within the polymer structure, which can coordinate stoichiometrically with Fe^{3+} ions [66]. To achieve optimal coagulant performance, it was anticipated that each –COOH group would combine with one Fe^{3+} ion, resulting in maximum positive charge density and active complexation sites for organic pollutants. The pH condition of the synthesis medium was crucial, considering the deprotonation of PAA and the hydrolysis of Fe^{3+} during the preparation process. In this study, the coagulant was synthesized by slowly adding a 0.14 mol/L sodium polyacrylate (PAAS) solution into a 0.185 mol/L $FeCl_3 \cdot 6H_2O$ solution under rapid stirring at 25 °C. To prevent precipitation, the pH of PAAS was adjusted to 3 before the titration step. The resulting coagulants, prepared with Fe^{3+} to PAA molar ratios of 1:0.1, 1:1, and 1:2, were labeled Fe-PAA-1:0.1, Fe-PAA-1:1, and Fe-PAA-1:2, respectively. The initial concentration of Fe was determined to be 0.1 mM. The pH condition is crucial in the coagulation process, as it affects the deprotonation of HA, hydrolysis of Fe^{3+}, and subsequent interaction between humic acid and the coagulant. As such, all the coagulants in this study demonstrated superior coagulation performance at pH 5 compared to pH 7 and 9, with regard to optimal TOC removal rates. The findings indicated that Fe-PAA-1:0.1 achieved around 80% TOC removal, Fe-PAA-1:1 exceeded 80% TOC removal, and Fe-PAA-1:2 achieved just under 80% TOC removal. Overall, the coagulant Fe-PAA-1:1 with an R ratio of 1:1 showed the best coagulation performance due to its broad applicable pH range and low residual Fe^{3+} level [66].

Composite coagulants, specifically PSiFAC (polyaluminum ferric silicate chloride), were synthesized at room temperature under various experimental conditions and different ratios of components and polymerization modes [67]. The initial solutions used included 0.5 M $AlCl_3 \cdot 6H_2O$, 0.5 M $FeCl_3 \cdot 6H_2O$, 0.5 M NaOH as the added base, and a prepared polysilicic acid (pSi) solution. The process involved adding an appropriate amount of ferric chloride solution to an aluminum chloride solution, with vigorous stirring, at specific Al/Fe ratios. Subsequently, the pSi was introduced at desired Al+Fe/Si ratios. Finally, the polymerization of aluminum was achieved by slowly adding the required amount of NaOH solution under magnetic stirring. Three different industrial wastewater samples were analyzed: tanner wastewater with 668 NTU turbidity, 6800 mg/L COD, 1.76 mg/L phosphates, and 2.981 UV_{254nm}; yeast manufacturing wastewater that had undergone preliminary anaerobic treatment, with 418 NTU turbidity, 11,455 mg/L COD, 3.49 mg/L phosphates, and 3.748 UV_{254nm}; the same yeast wastewater after subsequent aerobic treatment, showing 143 NTU turbidity, 4590 mg/L COD, 2.40 mg/L phosphates, and 3.307 UV_{254nm}. The addition of 80–100 mg/L of $PSiFAC_{1.5:10:15}$ achieved the following results: for tanner wastewater, 96% reduction in NTU turbidity, 67% reduction in COD, 62% reduction in phosphates, and 10% reduction in UV_{254nm}; for yeast manufacturing wastewater, 14% reduction in NTU turbidity, 22% reduction in COD, 38% reduction in phosphates, and 15% reduction in UV_{254nm}; and for yeast wastewater after aerobic treatment, 40% reduction in NTU turbidity, 56% reduction in COD, 43% reduction in phosphates, and 25% reduction in UV_{254nm} [67].

As per Tolkou et al. 2015 [67] $PSiFAC_{1.5:10:15}$, $PAFSiC_{1.5:15:10}$, $PFASiC_{1.5:15:10}$, and $PACl_{1.5}$ invested to remove turbidity and UV_{254nm} from simulated surface water (17.2 NTU and 0.153 UV_{254nm} absorbance) and from tanner wastewater (>2000 NTU). Fe solution was added to Al solution with vigorous stirring to achieve desired Al/Fe ratios, forming pFA. To prepare coagulants, pSi solution was mixed with pFA at desired Al+Fe/Si ratios, and a base solution was slowly added under magnetic stirring to reach desired OH/Al ratios. Coagulants made using co-polymerization are called PSiFAC. $PSiFAC_{1.5:10:15}$ denotes OH/Al 1.5, Fe/Si 10, and Al/Si+Fe 15. For PAFSiC, pFSi was mixed with an Al solution at various Al/Si+Fe ratios, and a base solution was slowly added to reach desired OH/Al ratios. Coagulants made by co-polymerization are called PAFSiC. $PAFSiC_{1.5:15:10}$ signifies OH/Al 1.5, Fe/Si 15, and Al/Si+Fe 10. Similarly, pSi solution was stirred into Al solution to achieve desired Al/Si ratios for each reagent, resulting in pASi for coagulant preparation. PFASiC, produced via co-polymerization, follows OH/Al 1.5, Fe/Si 15, and Al+Fe/Si 10 ($PFASiC_{1.5:15:10}$). Polyaluminum chloride solutions (PACl-18) were also prepared under identical conditions for comparison, excluding the inclusion of silicates and ferric compounds with OH/Al 1.2. The characterization of simulated surface water was turbidity 17.2 NTU and absorbance UV_{254nm} 0.153. Otherwise, the tanner wastewater had before treatment > 2000 NTU turbidity. Coagulant concentrations ranged from 2–3 mg/L for simulated surface water and 100 mg/L for tanner wastewater, with experiments conducted at pH 7. $PSiFAC_{1.5:10:15}$ achieved a 97% removal rate, while $PACl_{1.5}$ showed the highest removal rate of 93% at a dose of 2 mg/L. Similarly, for UV254nm absorbance reduction, $PSiFAC_{1.5:10:15}$ was the most effective, followed by $PFASiC_{1.5:15:10}$, especially at concentrations above 2–3 mg/L. Coagulants prepared by composite polymerization were less efficient, as observed in turbidity results. However, $PSiFAC_{1.5:10:15}$ achieved a 93% UV reduction rate, compared to 78% with $PACl_{1.5}$. The top-performing coagulants, $PSiFAC_{1.5:10:15}$, $PAFSiC_{1.5:15:10}$, and $PFASiC_{1.5:15:10}$, were used to treat tanner wastewater, significantly reducing turbidity (>99.6%) and absorbance at doses exceeding 100 mg/L [67].

An innovative coagulant, poly–aluminum–ferric–silicate–chloride ($PAPEFAC_{1.5\text{-}10\text{-}15}$) was used to eliminate turbidity, COD, and phosphates from the same three different wastewater, referred above [68]. $FeCl_3$ solution were added $AlCl_3$ solution under vigorous stirring. Next, anionic polyelectrolyte (APE) solution were added, followed by the slow addition of base solution. The mixture was stirred for 1 h to mature and then diluted with water to a final concentration of 0.1 M relative to Al. $PAPEFAC_{1.5\text{-}10\text{-}15}$ indicates OH/Al:1.5, Al/Fe:10, Al+Fe/APE:15. The removal efficiencies for tanner wastewater were approximately 96% for turbidity, 65% for COD, and less than 65% for phosphates. For yeast wastewater after anaerobic treatment, the removal efficiencies were about 20% for turbidity, 5% for COD, and less than 30% for phosphates. Following additional post-aerobic treatment, yeast wastewater showed removal efficiencies of around 40% for turbidity, 15% for COD, and 57% for phosphates [68].

Two composite inorganic pre-polymerized coagulants were synthesized: PSi-FAC-$Na_{1.5\text{-}10\text{-}15}$ (polyaluminum ferric silicate chloride) and PSiFAC-$Mg_{30\text{-}10\text{-}15}$ (polyaluminum ferric silicate magnesium) [69]. For the first coagulant, $FeCl_3$ solution was added to $AlCl_3$ solution under vigorous stirring at ratio Al/Fe 10. Then, the pSi solution was introduced to the resulting Fe/Al solution at Al+Fe/Si 15 ratio, followed by the gradual addition of an alkaline solution (NaOH) until reaching the desired pH value (3–3.50) and Al/OH 1.5. Similarly, for the second coagulant, $FeCl_3$ solution was added to an $AlCl_3$ solution under vigorous stirring, followed by the addition of pSi solution. Subsequently, a MgO slurry was slowly added to the mixture until the desired pH value (2–2.5) was achieved. PSiFAC-$Mg_{30\text{-}10\text{-}15}$ signifies OH/Al 30, Al/Fe 10, and Al+Fe/Si 15. With an initial fluoride concentration of 5 mg F/L and a coagulant dose of 30 mg Al/L, the uptake capacity (Q1.5-values) to achieve a residual fluoride concentration of 1.5 mg F/L at pH 7.0 was found to be 170 mg F/g Al for PSiFAC-$Mg_{30\text{-}10\text{-}15}$ and 94 mg F/g Al for PSiFAC-$Na_{1.5\text{-}10\text{-}15}$ [69].

In Table 3, all of the novel synthetic coagulants presented and compared in removal rate in Figure 3. The best removal rates for turbidity had PALS coagulants with approximately 99% [64], after $PSiFAC_{1.5:10:15}$ in simulated wastewater (97%) [67], and $PAPEFAC_{1.5\text{-}10\text{-}15}$ (~96%) [68]. POFC-2 coagulant demonstrated the highest efficiency in removing TSS from domestic wastewater, achieving up to 93% removal and also achieving up to 89% COD removal in domestic wastewater [19].

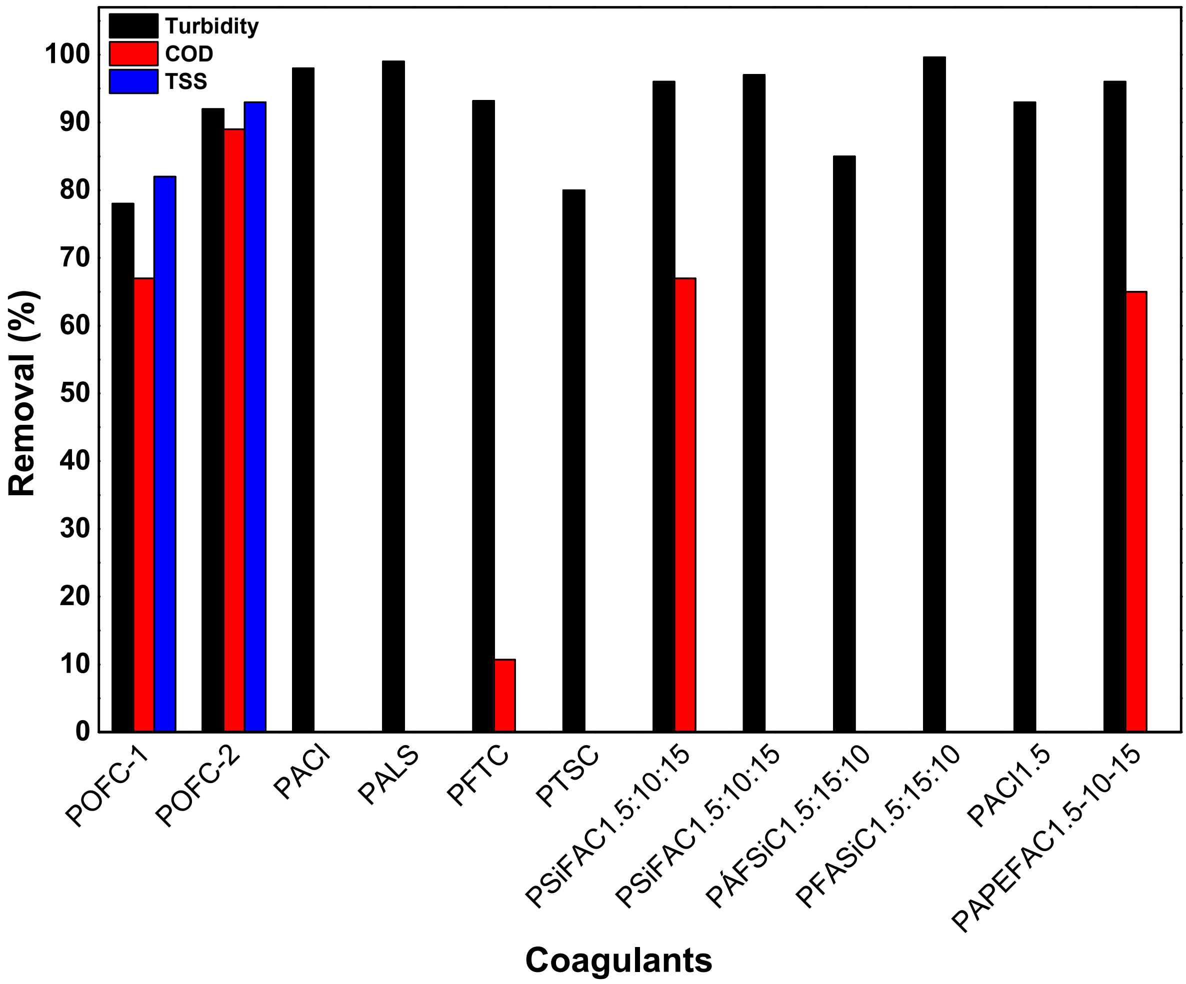

Figure 3. Comparison of novel inorganic coagulants as per removal rate for turbidity, COD, and TSS.

Table 3. Inorganic novel coagulants for wastewater treatment.

Coagulants	Remove	Initial Conditions	pH	Coagulant Dosage	Removal	Ref.
POFC-1 [1] POFC-2 [2]	Domestic Wastewater	86 NTU turbidity 342 mg/L COD 300 mg/L TSS	6–8	10 mg/L	78.00% turbidity 67.00% COD 82.00% TSS 92.00% turbidity 89.00% COD 93.00% TSS	[19]
PFS [3]	printing and dyeing wastewater	1 mg/L Sb 0.085 mg/L Sb(III), and 0.1 mg/L Sb(V)	5	60 mg/L	95.00% Sb(III), 90.00% Sb(V) 97.50% total Sb	[27]
$FeCl_3$–$AlCl_3$ [4] $FeSO_4$–$Al_2(SO_4)_3$ [5]	–	0.519 mg/L Total P	5 7	21.85 mg/L 15 mg/L	91.31% Total P 86.82% Total P	[61]
PASS [6]	Coking Wastewater	196.67 mg/L COD_{Cr}	7.0	7 mmol/L	69.50% COD_{Cr}	[62]
PACl [7]	Domestic Wastewater	270 NTU turbidity 100 mg/L DOC	6.5	25 mg/L	98.00% turbidity 69.80% DOC	[63]
PALS [8]	Simulate Wastewater	28.6–30.2 NTU Turbidity 2 mg/L total phosphorus 29.68–30.42 mg/L DOC	8 6 3–7	12 mg/L 8.0 mg/L 14 mg/L	99.00% turbidity 99.60% phosphate 69.57% DOC	[64]
PFTC [9] PTSC [10]	Coking Wastewater	40 NTU turbidity 480 mg/L DOC 2000 mg/L COD	9	800 mg/L	93.20% turbidity 10.10% DOC 10.70% COD 80.00% turbidity	[65]
Fe-PAA-1:0.1 [11] Fe-PAA-1:1 [12] Fe-PAA-1:2 [13]	Humid acid	TOC	5.0	0.1 mM Fe	~80.00% TOC >80.00% TOC <80.00% TOC	[66]
$PSiFAC_{1.5:10:15}$ [14]	Tanner Wastewater Yeast wastewater after aerobic treatment Yeast manufacturing wastewater without preliminary anaerobic treatment	668 NTU turbidity 6800 mg/L COD 1.76 mg/L phosphates 2.981 UV_{254nm} 418 NTU turbidity 11455 mg/L COD 3.49 mg/L phosphates 3.748 UV_{254nm} 143 NTU turbidity 4590 mg/L COD 2.40 mg/L phosphates 3.307 UV_{254nm}	7.8	80	96.00% turbidity 67.00% COD 62.00% phosphates 10.00% UV_{254nm} 14.00% turbidity 22.00% COD 38.00% phosphates 15.00% UV_{254nm} 40.00% turbidity 56.00% COD 43.00% phosphates 25.00% UV_{254nm}	[67,68]
$PSiFAC_{1.5:10:15}$ [14] $PAFSiC_{1.5:15:10}$ [15] $PFASiC_{1.5:15:10}$ [16] $PACl_{1.5}$ [17]	Simulated surface water Tanner wastewater Simulated surface water Tanner wastewater Simulated surface water Tanner wastewater Simulated surface water	17.2 NTU turbidity 0.153 UV_{254nm} >2000 turbidity 17.2 NTU turbidity 0.153 UV_{254nm} >2000 turbidity 17.2 NTU turbidity 0.153 UV_{254nm} >2000 turbidity 17.2 NTU turbidity 0.153 UV_{254nm}	7.0	2 mg/L 3 mg/L 100 mg/L 2 mg/L 3 mg/L 100 mg/L 2 mg/L 3 mg/L 100 mg/L 2 mg/L 3 mg/L	97.00% turbidity 93.00% UV_{254nm} 99.60% turbidity ~95.00% UV_{254nm} ~85.00% turbidity 99.70% UV_{254nm} ~95.00% turbidity >90.00% UV_{254nm} 99.6.00% turbidity 93.00% turbidity 78.00% UV_{254nm}	[67]
$PAPEFAC_{1.5\text{-}10\text{-}15}$ [18]	Tanner Wastewater Yeast wastewater after aerobic treatment Yeast manufacturing wastewater without preliminary anaerobic treatment	668 NTU turbidity 6800 mg/L COD 1.76 mg/L phosphates 418 NTU turbidity 11455 mg/L COD 3.49 mg/L phosphates 143 NTU turbidity 4590 mg/L COD 2.40 mg/L phosphates	7.8	80 mg Al/L	~96.00% turbidity ~65.00% COD <65.00% phosphates ~20.00% turbidity ~5.00% COD <30.00% phosphates ~40.00% turbidity ~15.00% COD ~57.00% phosphates	[68]
$PSiFAC\text{-}Mg_{30\text{-}10\text{-}15}$ [19] $PSiFAC\text{-}Na_{1.5\text{-}10\text{-}15}$ [20]	Industrial Wastewater	5 mg F/L fluoride	7.0	30 mg Al/L	Q1.5 170 mg F/g Al Q1.5 94 mg F/g Al	[69]

[1,2] poly-ferric chloride OH/Fe:1 or 2, [3] polymeric ferric sulfate, [4] ferric chloride with aluminum chloride, [5] ferrous sulfate with aluminum sulfate, [6] polyaluminum silicate sulfate, [7] polyaluminum chloride, [8] polyaluminum Lanthanum Silicate, [9] poly-ferric-titanium-silicate-chloride, [10] poly-titanium-silicate-chloride, [11] Fe^{3+} to polyacrylic acid molar ratios of 1:0.1, [12] Fe^{3+} to polyacrylic acid molar ratios of 1:1, [13] Fe^{3+} to polyacrylic acid molar ratios of 1:2, [14,15,16] polyaluminum ferric silicate chloride, [1,16] polyaluminum ferric silicate chloride, [17] polyaluminum chloride, [18] poly–aluminum–ferric–APE–chloride, [19] polyaluminum ferric silicate magnesium, [20] polyaluminum ferric silicate chloride.

Inorganic Coagulants with Flocculants Added

Chitosan powder was mixed with HCl solution and stirred until fully dissolved, after which the solution was diluted with distilled water [70]. Vegetable oil refinery wastewater (VORW) contains substantial refractory pollutants that require effective treatment before discharge. This study examines the treatment of VORW using coagulation-flocculation with ferric chloride ($FeCl_3$) as the coagulant and chitosan as the natural flocculant. The removal efficiencies of 3753 NTU turbidity, 7680 mg/L COD, and polyphenols were evaluated. The results indicated that the optimal treatment conditions were a pH of 6, $FeCl_3$ dosage of 1.6 g/L, chitosan dosage of 13.4 mg/L, and an agitation time of 26 min. Under these conditions, 100% turbidity removal, 86% COD reduction, and 90% polyphenol removal were achieved. In the coagulation-flocculation process employed in this study, $FeCl_3$ serves as a coagulant by destabilization of colloidal particles, thereby promoting their aggregation into bigger aggregates. Conversely, chitosan acts as a flocculant, linking particles together to form flocs. The amino groups in chitosan interface with particles via hydrogen bonding, and electrostatic attraction, contributing to the formation of denser and bigger flocs that settle rapidly [70].

In the Table 4 are presented the inorganic coagulant $FeCl_3$ with an organic flocculant chitosan in removing from vegetable oil refinery wastewater turbidity, COD, and polyphenols [70].

Table 4. Novel inorganic coagulants with addition of flocculants for wastewater treatment.

Coagulant	Flocculant	Remove	Initial Conditions	pH	Coagulant Dosage	Flocculant Dosage	Removal	Ref.
$FeCl_3$ [1]	chitosan	Vegetable oil refinery wastewater	3753 NTU turbidity 7680 mg/L COD 168.36 mg/L polyphenols	6.0	1.6 g/L	13.4 mg/L	100.00% turbidity 86.00% COD 90.00% polyphenol	[70]

[1] $FeCl_3$: Ferric chloride.

4. Organic Coagulants

Chitosan obtained from shrimp shells and having a deacetylation degree of 75% and higher, was employed to treat brewery wastewater with a turbidity of 160 NTU, TOC concentration of 176 mg/L, and an orthophosphate level of 139 mg/L [71]. Under optimal conditions of pH at 8, a dose of 2 g/L, and a contact time of 43 min, the experiments achieved removals of 91% for turbidity, also, 89% for TOC, and for orthophosphates 65% [71].

The cetyltrimethylammonium bromide (CTAB) was purchased [72]. Coagulation/flocculation experiments were carried out with initial concentrations of Cr(III), Ni(II), Cu(II), Zn(II), and Cd(II) set at 10.4 mg/L, a coagulant dose of 57 mg/L, and a pH value of 10.5. The removal percentages for Cr, Ni, Cu, Zn, and Cd using the CTAB coagulation method are approximately 98%, 92.69%, 96.63%, 99.35%, and 99.52%, respectively [72].

Epichlorohydrin was mixed with diphenylamine at specified epichlorohydrin-diphenylamine molar ratios of 1:1, 1.5:1, 2:1, and 2.5:1 for PE-1, PE-2, PE-3, and PE-4, respectively, by gradual addition into the reactor with continuous stirring to initiate oligomer formation [73]. Following this, 1,2-diaminoethane was incorporated into the mixture at a selected weight percentage (1%) with ongoing stirring. The reaction mixture was maintained at a temperature range of 60–80 °C. After eight hours, the polyamine polymers were obtained in solid form, as depicted in Figure 4 [73].

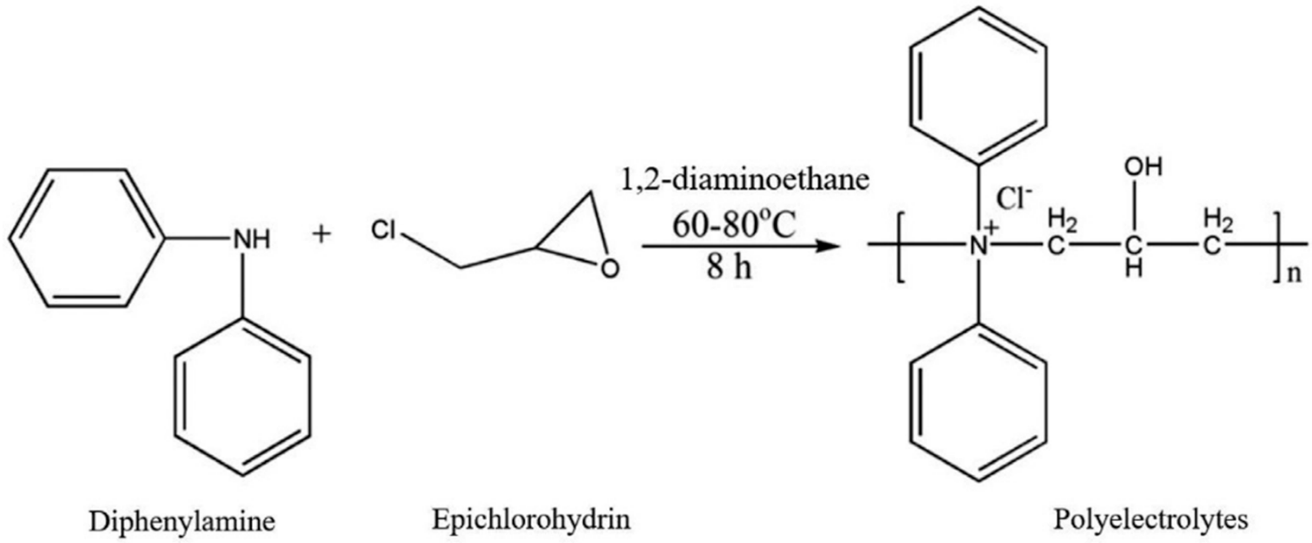

Figure 4. Polyelectrolyte synthesis [73].

The sugar industry wastewater displayed turbidity levels of 83 NTU, with total suspended solids (TSS) measured at 1120 mg/L, chemical oxygen demand (COD) at 991 mg/L, biochemical oxygen demand (BOD) at 543 mg/L, and a pH value of 6.1. PE-2 demonstrated superior efficacy in removing turbidity compared to other polyelectrolytes. At a dosage of 1.0 mg/L, PE-2 achieved an outstanding result with residual turbidity as low as 1 NTU. In contrast, PE-3 exhibited a lower performance, removing only 87% of turbidity at the same dosage. PE-4 and PE-1 followed in efficacy, with turbidity removal rates of 78.50% and 74.68%, respectively, at a dosage of 2.0 mg/L. Similarly, the sequence of TSS removal mirrors that of turbidity. PE-2 achieved a remarkable 98.40% removal of TSS at a mere dosage of 1.0 mg/L. Meanwhile, TSS removal rates stood at 70.25%, 86.59%, and 75.20%, and for PE-1, PE-3, and PE-4, respectively. PE-2 demonstrated superior efficacy in removing turbidity compared to other polyelectrolytes. At a dosage of 1.0 mg/L, PE-2 achieved an outstanding result with 100% turbidity removal. In contrast, PE-3 exhibited a lower performance, removing only 87% of turbidity at the same dosage. PE-1 and PE-4 followed in efficacy, with turbidity removal rates of 74.68% and 78.50%, respectively, at a dosage of 2.0 mg/L. The removal of COD and BOD follows the sequence of PE-2 > PE-3 > PE-1 > PE-4, indicating that PE-2 exhibits the highest efficiency in removing both pollutants. At a dosage of 1.5 mg/L, PE-2 achieved remarkable removal rates of 94.5% for COD and 95.2% for BOD. In comparison, PE-3 removed 82.25% of COD and 79.52% of BOD under similar conditions. However, the performance of PE-1 and PE-4 was less satisfactory, with COD removal rates of 72.48% and 65.36%, respectively, and BOD removal rates of 71.47% and 63.67%, respectively. This disparity in performance may be attributed to the lower charge density of PE-1 and PE-4. Typically, polyelectrolytes carry higher charges, leading to enhanced adsorption of suspended particles through electrostatic attraction between positively and negatively charged ionic groups. The removal pattern of oil, and grease mirrors that of COD and BOD [73].

Table 5 summarizes all novel organic coagulants such as chitosan, CTAB, PE-1, PE-2, PE-3, and PE-4 for wastewater treatment. Figure 5 makes a comparison was made regarding the effectiveness of novel organic coagulants in removing turbidity, BOD, COD, and TSS. PE-2 stands out for its effectiveness in removing turbidity, TSS, COD, and BOD from sugar industry wastewater [73], while chitosan is particularly effective for TOC and orthophosphate removal in brewery wastewater [71]. It is notable CTAB is highly effective for the removal of various toxic metal ions in wastewater [72].

Table 5. Organic coagulants for wastewater treatment.

Coagulants	Remove	Initial Conditions	pH	Coagulant Dosage (mg/L)	Removal	Ref.
Chitosan	Brewery Wastewater	160 NTU turbidity 176 mg/L TOC 139 mg/L orthophosphate	8	2000	91.00% turbidity 89.00% TOC 65.00% orthophosphate	[71]
CTAB [1]	Toxic Metal−Organic Complexes	10.4 mg/L Cr(III) 10.4 mg/L Ni(II) 10.4 mg/L Cu(II) 10.4 mg/L Zn(II) 10.4 mg/L Cd(II)	10.5	57	98.00% Cr(III) 92.69% Ni(II) 96.63% Cu(II) 99.35% Zn(II) 99.52% Cd(II)	[72]
PE-1 [2]	Sugar industry wastewater	83 NTU Turbidity	6.1	2.0 1.0 1.5 1.5	74.68% turbidity 70.25% TSS 72.48% COD 71.47% BOD	[73]
PE-2 [3]		1120 mg/L TSS		1.0 1.0 1.5 1.5	100% turbidity 98.40% TSS 94.50% COD 71.47% BOD	
PE-3 [4]		991 mg/L COD		1.0 1.0 1.5 1.5	87.00% turbidity 86.59% TSS 82.25% COD 79.52% BOD	
PE-4 [5]		543 mg/L BOD		2.0 1.0 1.5 1.5	78.50% turbidity 75.20% TSS 65.36% COD 63.67% BOD	

[1] cetyltrimethylammonium bromide, [2] epichlorohydrin-diphenylamine 1:1, [3] epichlorohydrin-diphenylamine, [4] epichlorohydrin-diphenylamine 2:1, [5] epichlorohydrin-diphenylamine 2.5:1.

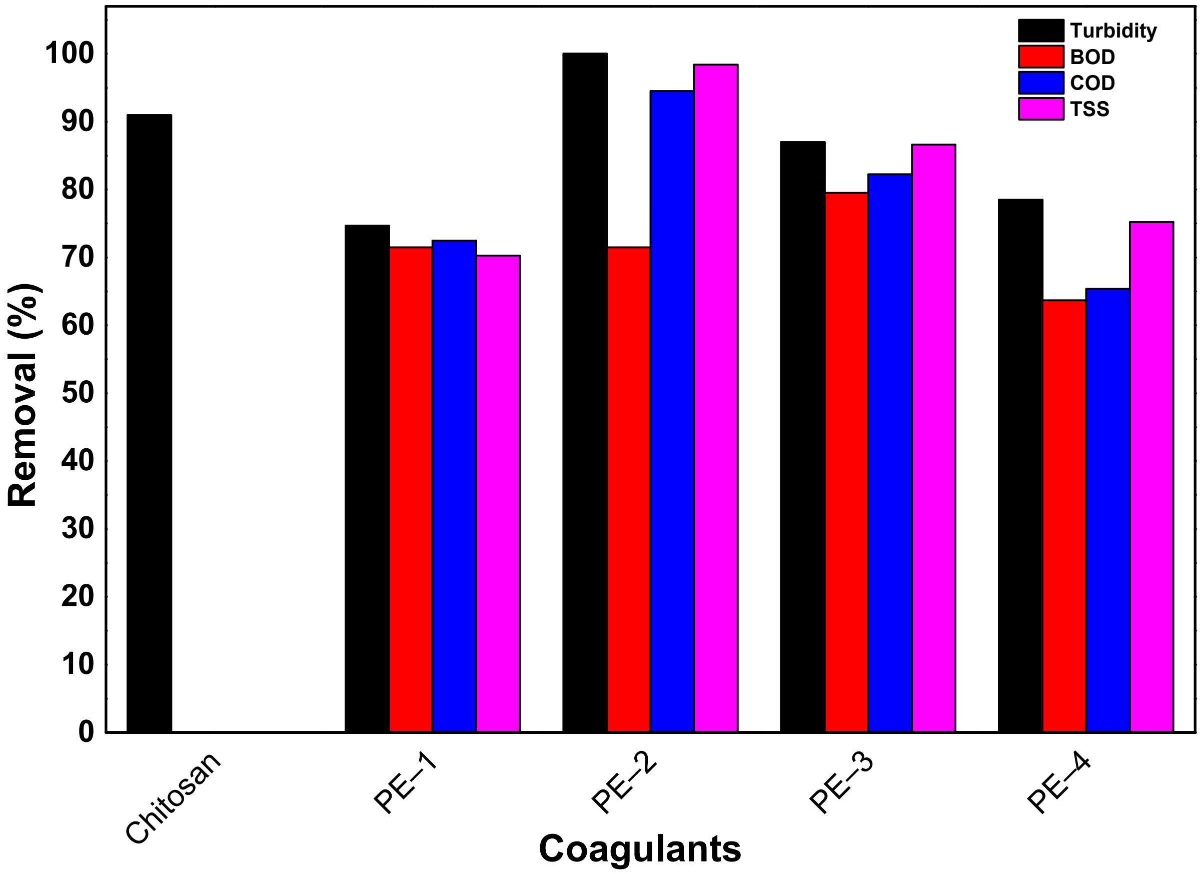

Figure 5. Comparison of novel organic coagulants as per removal rate for turbidity, BOD, COD, and TSS.

5. Hybrid Coagulants/Flocculants

3-aminopropyltriethoxysilane was used as the Si source to prepare organic silica hybrid coagulants (PAAP) [74]. To obtain defined Si/Al molar ratios of 0.05, 0.1, 0.2, 0.4, and 0.8, a mixture of 3-aminopropyltriethoxysilane, $AlCl_3$ solution, and water was prepared. Subsequently, NaOH solution was gradually introduced with continuous stirring to achieve the desired B values (basicity, OH/Al molar ratios) of 0, 0.5, 1, 1.5, and 2.0. After aging for 24 h, ethanol was removed by vacuum rotary evaporation at 30 °C. The coagulant solutions were then diluted to the same final Al concentration. The hybrid coagulants were labeled PAAP, with designations like $PAAP_{0.1,0.5}$ indicating a Si/Al ratio of 0.1 and a B value of 0.5. The two types of wastewaters were raw coking wastewater, characterized by pH 9.98, 105 NTU turbidity, 4654 mg/L COD, 1138.45 mg/L DOC, and 22.75 UV_{254nm}, and biologically treated coking wastewater, characterized by pH value of 8.13, 128 NTU turbidity, 109.03 mg/L COD, 41.86 mg/L DOC, and 2.197 UV_{254nm}. The optimal dosage for treating raw coking wastewater was determined to be 600 mg/L. At this dosage, the removal efficiencies were 82.05% for turbidity, 12.09% for UV_{254nm}, 9.34% for DOC, and 24.16% for COD. The optimal dosage for treating biologically treated coking wastewater was determined to be 300 mg/L. At this dosage, the removal efficiency for turbidity was 97.84%, for UV_{254nm} it was 37.72%, for DOC it was 29.97%, and for COD it was 63.39% [74].

Various inorganic-organic hybrids were prepared with PANS as the base product. PANS is a commercial polyaluminum nitrate sulfate featuring an intermediate aluminum content of 5.5% and a basicity of 46%., containing 16.0% NO_3 and 3.0% SO_4^{2-}. The hybrids were created by directly blending PANS with three different polyamines, each varying in molecular weight, at room temperature. The molecular weights of the polyamines followed the order: PA1 < PA2 < PA3. Four different addition levels were tested for each type of polyamine: 5%, 10%, 15%, and 20% by weight of commercial solutions (48–52% dry solids), with the remainder being PANS. Higher polyamine contents were also prepared, but these blends proved unstable, as indicated by the appearance of turbidity within 24 h of preparation. Additionally, PANS was tested alone to assess its efficiency relative to the hybrid materials. The hybrids are denoted as PANS-PAx-Z, where PAx represents the specific polyamine (PA1, PA2, or PA3) and Z indicates the polyamine weight content in the hybrid. The hybrid coagulants were categorized by their aluminum content and cationic charge according to the concentration of the commercial polyamine solution used. All the hybrids and PANS were tested together with the same flocculant: an anionic polyacrylamide with high molecular weight and medium charge. The effluent undergoes a treatment process that includes primary treatment via dissolved air flotation, followed by secondary treatment in a moving bed bioreactor, and another round of dissolved air flotation. Water samples from the final effluent, taken before discharge to an urban wastewater treatment plant, showed the following characteristics: raw water had a turbidity of 11.4 NTU, total solids concentration of 1990 mg/L, cationic demand of 0.52 meq/L, and in the dissolved fraction, total solids were 1890 mg/L. Additionally, the chemical oxygen demand (COD) was 256 mg/L, silica (SiO_2) content was 145 mg/L, calcium concentration was 33.7 mg/L, and magnesium concentration was 2.8 mg/L. The efficacy of various PANS-PA1 hybrids in silica removal varied slightly, achieving maximum removal rates of approximately 47% with PANS-PA1-20 at a dosage of 2500 mg/L. Similar efficiencies around 40% were observed with other PA1 hybrids at different dosages: 1000 mg/L for PANS-PA1-5, 1500 mg/L for PANS-PA1-10, and 2000 mg/L for PANS-PA1-15. Under initial pH conditions of 8.4, PANS-PA2 hybrids demonstrated slightly higher silica removal efficiencies compared to PANS-PA1, reaching about 50% with PANS-PA2-5 and PANS-PA2-10 at 2500 mg/L. At a lower dosage of 500 mg/L, silica removal ranged from 39% with PANS-PA2-5 to 23% with PANS-PA2-20, showing significant enhancement compared to PANS alone. Silica removal efficiencies with PANS-PA3 hybrids were slightly lower than with PA2 but higher than with PA1 hybrids. Maximum removal rates ranged from 51% with PANS-PA3-5 to 40% with PANS-PA3-20 at a dosage of 2500 mg/L. Notably, PANS-PA3-5 achieved 42% silica removal at 500 mg/L, compared to only 12% with PANS alone at the same dosage. Conversely, removal rates were lower with 20% PA3 compared to the other hybrids. At an initial pH of 10.5, silica removal increased consistently with coagulant dosage due to increased ionization of silica and greater availability of alkalinity for aluminum hydroxide formation, which are active species in coagulation. Differences in silica removal rates between hybrids and PANS were less pronounced at pH 10.5 compared to pH 8.4. Molecular weight of polyamine or percentage in the hybrids showed no significant effect. PANS alone achieved high removal rates, including 90% at 2500 mg/L and 71% and 83% at lower dosages of 1500 mg/L and 2000 mg/L, respectively. For hybrids, removal rates ranged approximately from 50% to 90% across various dosages tested. At 500 mg/L, removal efficiency varied notably, from 16% with PANS-PA3-20 to 31% with PANS-PA2-5, whereas PANS alone achieved 14% removal [75].

Initially, an HCl solution (10–30% *w*/*w*) and an H_2SO_4 solution (10–20% *w*/*w*) were added to a conical flask containing galvanized aluminum slag, which had been crushed into smaller, irregular pieces. This mixture was subjected to leaching for 1–5 h in a water bath constant temperature oscillator at 70–90 °C to obtain a leachate. The leachate was then filtered at 30–40 °C to produce a colorless filtrate. Subsequently, a specific concentration of PAM solution was added to the colorless filtrate either before or after polymerization. Following this, an NaOH solution (0.5–5.0% *w*/*w*) was slowly introduced into the mixture to facilitate polymerization for 2–48 h at 60–80 °C with continuous stirring, resulting in a liquid product of PPAZF with a pH of 2.5–3.5 [76]. This liquid PPAZF was then used in subsequent coagulation tests. The quality indicators for domestic sewage were as follows: turbidity was 232 NTU, COD was 661.51 mg/L, and pH was 7.71. In contrast, the quality indicators for pharmaceutical wastewater were: turbidity was 265 NTU, COD was 10,025 mg/L, and pH was 4.84. The rapid mixing was carried out at 200 rpm for 1 min. This was followed by a slow mixing stage at 60 rpm for 10 min to form dense and large microflocs. Finally, a sedimentation stage was conducted for 15 min. The removal achieved with initial coagulant dosage 148 mg/L (Al concentration). For domestic sewage, the removal efficiencies were approximately 98% for turbidity and 96% for COD. For pharmaceutical wastewater, the removal efficiencies were about 73% for turbidity and 47% for COD. Throughout the coagulation process, four mechanisms contribute significantly: electrical neutralization, compressed double electric layer, adsorption bridging, and sweep function. Specifically, the coagulation mechanism of PPAZF involves not only the binding and electrical neutralization of impurities and hydrolyzed forms of aluminum, zinc, and iron, but also entails the formation of complex precipitates due to the hydrolysis products of metals interacting with organic matter [76].

In Table 6 are illustrated the hybrid coagulants used for wastewater treatment. PAAP achieved significant turbidity removal for both raw and biologically treated coking wastewaters, with removal rates up to 97.84% and 82.05% respectively, compared to PANS-PAx hybrids which reached up to 90% and 50% silica removal at optimal dosages [75,76]. Domestic sewage showed higher removal rates with PPAZF (98% turbidity, 96% COD) compared to pharmaceutical wastewater (73% turbidity, 47% COD), indicating varying treatment requirements based on wastewater composition and characteristics [76].

Table 6. Hybrid coagulants for wastewater treatment.

Coagulants	Remove	Initial Conditions	pH	Coagulant Dosage (mg/L)	Removal	Ref.
PAAP$_{0.1,0.5}$ [1]	Raw coking wastewater	105 NTU turbidity 4654 mg/L COD 1138.45 mg/L DOC 22.75 UV$_{254nm}$	9.98	600	82.05% turbidity 24.16% COD 9.34% DOC 12.09% UV$_{254nm}$	[74]
	Biologically treated coking wastewater	128 NTU turbidity 109.03 mg/L COD 41.86 mg/L DOC 2.197 UV$_{254nm}$	8.13	300	97.84% turbidity 63.39% COD 29.97% DOC 37.72% UV$_{254nm}$	
PANS [2]	Raw water	11.4 NTU turbidity	10.5	500	14.00% silica	[75]
				1500	71.00% silica	
				2000	83.00% silica	
		1990 mg/L Total Solids (raw water)		2500	90.00% silica	
PANS-PA1-Z [3]				500	~86.00% silica	
				1000	50.00% silica	
				1500	70.00% silica	
		0.52 meq/L Cationic Demand		2000	80.00% silica	
				2500	90.00% silica	
PANS-PA2-Z [4]		1890 mg/L TOTAL Solids (dissolved fraction)		500	31.00% silica	
				1000	50.00% silica	
				1500	70.00% silica	
				2000	80.00% silica	
		256 mg/L COD		2500	90.00% silica	
PANS-PA3-Z [5]				500	16.00% silica	
				1000	50.00% silica	
				1500	70.00% silica	
		145 mg/L SiO_2 Silica		2000	80.00% silica	
				2500	90.00% silica	
PANS-PA1-5 [6]	Dissolved Fraction	33.7 mg/L Calcium	8.4	1000	~40.00% silica	
PANS-PA1-10 [7]				1500	~40.00% silica	
PANS-PA1-15 [8]				2000	~40.00% silica	
PANS-PA1-20 [9]				2500	47.00% silica	
PANS-PA2-5 [10]				500	39.00% silica	
				2500	~50.00% silica	
PANS-PA2-10 [11]				500	35.00% silica	
				2500	~50.00% silica	
PANS-PA2-15 [12]		2.8 mg/L Magnesium		500	26.00% silica	
PANS-PA2-20 [13]				500	23.00% silica	
				2500	40.00% silica	
PANS-PA3-5 [14]				500	42.00% silica	
				2500	51.00% silica	
PANS-PA3-20 [15]				2500	40.00% silica	
PPAZF [16]	Domestic sewage	232 NTU turbidity 661.51 mg/L COD	7.71	148	~98% turbidity ~96% COD	[76]
	Pharmaceutical wastewater	265 NTU turbidity 10,025 mg/L COD	4.48		~73% turbidity ~47% COD	

[1] Si/Al:0.1, OH/Al: 0.5, [2–15] polyaluminum nitrate sulfate/polyamine [16] poly-polyacrylamide-Al-Zn-Fe.

6. Conclusions

In summary, coagulation/flocculation are processes aimed at efficiently removing turbidity, total suspended solids (TSS), chemical oxygen demand (COD), biochemical oxygen demand (BOD), toxic metals, phosphates, and UV_{254nm} from wastewater. Natural, inorganic, and organic coagulants can be utilized either independently or in combination with flocculants.

Among natural coagulants, Crab Shell Bio-Coagulant (CS) emerges as the most effective in turbidity removal, achieving a remarkable 98.91% removal rate. Oak leaves protein exhibits superior performance in removing TSS and COD concentrations.

In contrast, synthetic inorganic coagulants like PALS, $PSiFAC_{1.5:10:15}$, and $PAPEFAC_{1.5\text{-}10\text{-}15}$ demonstrate exceptional turbidity removal rates, reaching approximately 99%, 97%, and 96% respectively. POFC-2 coagulant stands out for its efficiency in removing TSS and COD from domestic wastewater, achieving up to 93% removal for TSS and 89% for COD.

Additionally, the combination of $FeCl_3$ as an inorganic coagulant and chitosan as an organic flocculant shows promise in removing turbidity, COD, and polyphenols from vegetable oil refinery wastewater.

Among the novel organic coagulants, PE-2 exhibits remarkable effectiveness in removing turbidity, TSS, COD, and BOD from sugar industry wastewater, while chitosan demonstrates efficacy in TOC and orthophosphate removal in brewery wastewater. CTAB proves highly effective in removing various toxic metal ions from wastewater.

Furthermore, the hybrid coagulants $PAAP_{0.1,0.5}$ and PPAZF achieve outstanding removal of turbidity, approximately 98%. At pH 10.5, PANS-PA1, PANS-PA2, and PANS-PA3 demonstrated superior silica removal compared to their performance at pH 8.4.

Overall, these findings underscore the diverse applications and effectiveness of different coagulants in treating various types of wastewaters, offering valuable insights for wastewater treatment strategies.

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