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Hello,

Prayudhy Yushananta has submitted the manuscript, "Novel Copolymer Cationic from Agroindustrial Waste using Microwave" to Open Access Macedonian Journal of Medical Sciences.

If you have any questions, please contact me. Thank you for considering this journal as a venue for your work.

Prof. Dr Mirko Spiroski

Novel Copolymer Cationic from Agroindustrial Waste using Microwave

ABSTRACT

Background. A cationic copolymer has been developed as a substitute for synthetic coagulants, resulting in decreased pH, potential health problems, high costs, and large sludge volumes.
Aim. This study evaluated the potential of banana pith in several treatments as a natural coagulant to reduce turbidity, COD, and color.
Methods. The synthesis was carried out by inserting the cationic moiety of GTA (3-Chloro-2-hydroxypropyl trimethyl ammonium chloride) into the starch backbone by microwave radiation.
Resulth. It has been found that the floculation characteristics depend on the charge neutralization, followed by the linkage between the copolymer chains. The results showed that the initial dose and concentration influenced the copolymer's flocculation performance.
Conclusions. Natural polysaccharides can be modified becomes an effective flocculation material for treating clean water and wastewater.

Keywords: Coagulation-flocculation, banana pith, turbidity, COD, color

Introduction

Water supply is intended so that residents can carry out their activities as humans [1,2]. There are two serious problems in the provision of clean water, namely the increasing population and the rapid rate of urbanization [3–10]. It is estimated that the demand for water needs will increase drastically in the world from 9.7 billion in 2020 to 9.7 billion people in 2050 [11]. In the industrial sector, clean water is needed to reach about 20% of the total water available [3]. Problems in the availability aspect are the increase in water pollution from various toxic pollutants as a consequence of the rapidly growing economic development [6,7,9,12–16]. The main problems in industrial waste disposal are hydraulic overload, temperature extremes, amounts of oil and grease, acids or bases, suspended solids, inorganic or organic materials, toxic, and volatile, odor, or corrosive gases [6,7,9,12–16].

These two problems, encourage massive exploitation of groundwater [16,17], resulting in a reduced volume of groundwater storage, land subsidence, negative impacts on water supply, decreased surface water flow and loss of springs, and loss of wetlands thus threatening the sustainability of supply water [18,19]. Globally, it is estimated that there has been a reduction in groundwater during 1900-2008 of 4.500 km^3 or the equivalent of a sea-level rise of 12.6 mm (more than 6% of the total). Since about 1950 the rate of groundwater reduction has increased significantly with the maximum rate occurring during 2000-2008, averaging about 145 km^3/year, or equivalent to 0.40 mm/year of level increase or 13% of the reported rate was 3.1 mm/year during this last period [20] [18] .

Many technologies have been developed to treat water and wastewater. such as precipitation, adsorption, coagulation, flotation, ion exchange, membrane filtration, and biological and electrolytic methods have been used to remove particles from water [21–34]. However, coagulation and flocculation techniques are the most widely applied technology in the world as a vital step in removing colloid particles, natural organic matter, microorganisms, and inorganic ions present in untreated water [16,30,32,35–38].

A coagulant is a very important material in the coagulation-flocculation process which refers to the agglomeration process of colloidal particles with an average size of 5-200 nm and small suspended solids in water, influenced by several factors such as temperature, ionic strength, pH. type and dose of material, coagulant, size and distribution type, concentration and properties of organic matter and colloid particles in suspension [4,9,10,13,16,22,23,25,26,35,37–46].

Metal salts such as ferric chloride ($FeCl_3$), aluminum sulfate (Al_2 (SO_4) 3), Pollyalluminium Chloride (PAC), Polyferrous sulfate (PFS), and poly ferric chloride (PFC) have been widely used to treat water and various wastewater [6,13,35,47,48]. Despite providing great performance in water treatment, these coagulants have several disadvantages such as reducing pH to acid, inefficiency in low temperatures, potential causes of health problems (such as Alzheimer's disease, neurotoxic and carcinogenic), relatively high cost of coagulants, and large sludge volume, and some evidence that iron salts and PFS can accelerate pipe corrosion [6,12,14,27,35,44,49,50]. Also, the chlorination method used in large quantities due to the use of high coagulant doses can produce several by-products with long-term harmful effects [10,12].

An alternative is needed to overcome them, one of which is to use plant-based coagulants because they are not toxic or corrosive [7,10,12,45,47,51–54]. Several natural polyelectrolytes have been studied as coagulants or flocculants in water and wastewater treatment with varying results. including rice bagasse [55]. *Tamarindus indica* [56]. *Moringa aloleifera* [57]. *Manihot esculenta* [58]. *Cactus opuntia* [59]. *Dolichos lablab* [60]. The advantages of using organic polyelectrolytes among others, the use of a lower coagulant dose, a smaller increase in ionic load on treated water, a decrease in the aluminum content in treated water, and cost savings [30,61].

Agricultural waste that has not been widely studied as raw material for natural coagulants is banana stem waste. Even though it is very potential because of its abundant and sustainable post-harvest availability [16,30,35,36,62]. The main ingredients in banana pith are water. Fibe, and polysaccharides or starch. The composition of C, H, O as much as 33.2%; 0.49%; and 6.17% [62], other studies mention 32.3% carbon, 4.21% hydrogen, 1.46% nitrogen, 43.5% oxygen, and 0.86% sulfur [30].

Polysaccharides are usually modified by conventional redox grafting methods [22,63–65]. The use of microwave radiation can be used in starch modification based on the advantages of energy saving, and speed [66–68]. In this study, starch from banana pith was synthesized through a copolymerization process using the microwave method, and at the same time, its flocculation performance was studied. The aim of this study was to obtain a natural coagulant that is modified from agricultural waste to reduce turbidity, chemical oxygen demand (COD), and color.

Materials and Methods

This study was conducted using the Way Kuripan River water which is the source of raw water for drinking water treatment (Figure 1). The water sample was dosed with natural coagulant followed by rapid mixing, flocculation, and sedimentation. The efficiency of removing contaminants is calculated by reducing turbidity, chemical oxygen demand (COD), and color. Banana pith is obtained from cavendish *(Musa paradisiaca)* harvested in Central Lampung Regency, Lampung Province.

GTA (3-Chloro-2-hydroxypropyl trimethyl ammonium chloride), sodium hydroxide, and ethanol were obtained from Sigma Aldrich. All these reagents were of analytical grade and

were applied directly. All solutions were prepared by deionized water. An Electrulox (Model No. EMM20M38GW) domestic microwave oven was used for the experiment.

Water sampling

Samples were collected from the Way Kuripan River in pre-cleaned containers (Fig. 1). The samples were stored in a cool box and transported to the laboratory. Sampling using the Standard Method [69].

Figure 1. Sampling coordinates (-5.4373308.105.2515303)

Starch of banana pith

Banana pith cleaned and rinsed with clean water and cut into small pieces. Soak with 0.5% Sodium Metabisulfite ($Na_2S_2O_5$) solution for 15 minutes to stop the browning effect. Maceration (grated). Squeezed, and filtered (0.4 mm) was performed to separate the liquid from the pulp. Do sedation so that the starch settles, the water is removed. Rinse the sediment with distilled water to clean the starch from the fibers. Dry in the oven at 80^0C until dry. Milling and shifting to get banana pith starch flour. Storage of banana pith flour in a desiccator to avoid additional moisture content.

Synthesis of natural coagulant with microwave radiation

Typically. 1.50g GTA (3-Chloro-2-hydroxypropyl trimethyl ammonium chloride) was added to a 100 mL container containing 5 mL sodium hydroxide (0.10 g/mL) aqueous. The solution was stirred thoroughly for 10 min. 10.00 g of starch is added to the mixture. Stirring was continued for 30 minutes on a water bath at 50^0C. Then, microwave irradiation was performed. Microwave irradiation (480 watts) has for 5 minutes at regular intervals. The microwave irradiation cycle is carried out to prevent the temperature from rising above 50^0C. The reaction vessel and its contents were cooled to room temperature. The precipitate is left in the reaction vessel and washed (3 times) with ethanol. Dry in an oven at 50^0C for 6 hours to obtain cationic starch.

Coagulant-flocculation test

Coagulation and flocculation tests were performed on water samples from the Way Kuripan River using the jar test. 500 mL river water was transferred to a 1000 ml beaker glass and placed in a jar-testing device. An initial pH check was performed, and all experiments were carried out at room temperature.

The desired amount of coagulant (Table 1) was added to the sample water and stirred at three variations of speed (rotary per minute) and time (minute). Followed by slow stirring at 30 rpm for 15 min. The mixture was left for 30 minutes to settle. The formation of floc particles was observed and recorded. Samples were taken with a pipette from a depth of 2 cm to determine turbidity, COD, and color. All analyzes were performed in quadruplicate.

Tabel 1. Dosage coagulants tested

Dosage (mg/L)	Speed (rpm)	Time (min)
50	150	2
100	180	3
200	200	5
300		

Analysis water samples

The Mettler Toledo brand pH meter is used to measure the pH value of the sample. The turbidity test used a portable turbidimeter (Model HACH 2100P). The principle of measuring turbidity is the ratio of the light intensity that is scattered by the sample. The sample is placed into the turbidimeter and the turbidity value is shown in NTU units. The total percentage of turbidity removal is calculated:

$$\text{Total turbidity percentage removal} = \frac{A-B}{A} \times 100 \qquad (1)$$

where A is the initial water turbidity (NTU). B is the turbidity after treatment (NTU).

The COD test was measured using UV Spectrophotometer HACH model. Chemical oxygen demand (COD) refers to the amount of oxygen required to oxidize the organic compounds in a water sample to carbondioxide and water. COD percentage removal was calculated as follows:

$$\text{COD percentage removal} = \frac{A-B}{A} \times 100 \qquad (2)$$

where A is the COD of row water (mg/L), B is COD after treatment (mg/L).

Statistical analysis

The effect of dose on turbidity. COD. color was analyzed two-way ANOVA, using software SPSS 24.0. Mathematical model assumption for a design is :

$$Y_{ijk} = \mu + \beta_i + \tau_j + \gamma_k + (\beta\tau)_{ij} + (\beta\gamma)_{ik} + (\tau\gamma)_{jk} + (\beta\tau\gamma)_{ijk} + \varepsilon_{ijk} \qquad (3)$$

i = 1,2,3,4; j = 1,2,3; k = 1,2,3, Y_{ijk}= the response of i^{th} level factor A, j^{th} level factor B, and k^{th} level factor C; μ= Grand mean; β_i = the effect of the i^{th} level factor A; τ_j= the effect of the j^{th} level factor B; γ_k = the effect of the k^{th} level factor B; $(\beta\tau)_{ij}$ = the effect of the combination of i^{th} level factor A with j^{th} level factor B; $(\beta\gamma)_{ik}$ = the effect of the combination of i^{th} level factor A with k^{th} level factor C; $(\tau\gamma)_{jk}$ = the effect of the combination of j^{th} level factor B with k^{th} level factor C; $(\beta\tau\gamma)_{ijk}$ = the effect of the combination of i^{th} level factor A with j^{th} level factor B with k^{th} level factor C; ε_{ij} = Error.

Results and Discussion

Characteric of natural coagulant

The copolymer is synthesized with microwave power (480 watts). The mechanism of microwave-assisted grafting is as follows: The microwave radiation rotates the GTA molecules, leading to the severing of its epoxypropyl bond, and the pi bond electron cloud splits up into two localized clouds (i.e., free radical sites on the constituent carbon atoms) [66,67]. Moreover, the polar groups absorb the microwave (e.g., OH groups attached to the starch molecule). Then behave as if they were anchored to an immobile raft. and its immobile localized rotations will occur in the microwave region [66], which eventually leads to the severing of these bonds, leading to the formation of free radical sites. The free radical sites created on the starch backbone and the GTA interact to yield the cationic graft copolymer through the usual free radical reaction mechanism.

Table 2 shows the high levels of amylopectin in banana pith starch. The grafting of positive ions into amylopectin will produce a more effective polymer because of the dangling polymer chains and highly branched structure [66,70]. The results of the synthesis gave a decrease in amylopectin by 9.87%. Indicates the occurrence of polymer chain termination due to microwave irradiation.

Table 2. Characteristics of banana pith starch

Characteristic	Starch (%)	Amylosa (%)	Amylopectin (%)
Row Starch	47.57	22.95	77.05
Syntesis starch	36.46	30.56	69.44

Treatment of water using natural coagulant

In general, the water quality of the Way Kuripan river shows that it does not meet the quality requirements for drinking water sources (Table 3). Water has significant turbidity, chemical oxygen demand (COD), and color values. Coagulation and flocculation of river water with natural coagulant showed a decrease of 94.4, 19.4, 87.5% of turbidity, COD, and color (Table 4). Even though the quality standards of clean water and drinking water have been achieved, the use of natural coagulants has significantly reduced turbidity, COD, and color.

Table 3. Characteristics of banana pith starch

Parameters	Row water	Treated water	Removal (%)	Guidelines for	
				Clean water	Drinking water
Turbidity (NTU)	563.0	31.4	94.4	25	5
COD (mg/L)	43.2	32.8	19.4	-	-
Color (TCU)	1900.0	238.2	87.5	50	15

Effect of natural coagulant on the removal turbidity. COD. and color

Table 4 shows the significant effect of dose on the decrease in turbidity and color ($p<0.05$), in contrast to the decrease in COD ($P>0.05$). Similarly, the effect of stirring speed and stirring time also significantly affected turbidity and color ($P<0.05$).

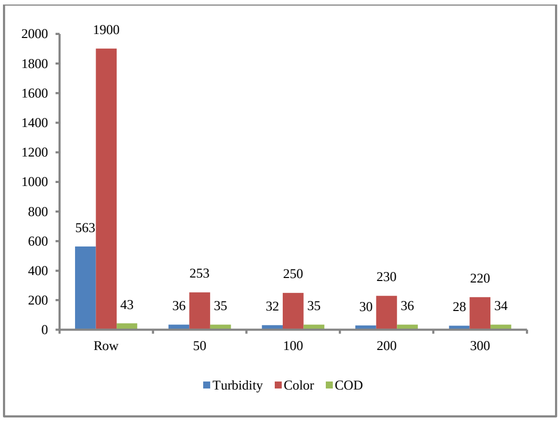
The combination of treatments was also observed in the experiment. A significant effect was found in all treatment combinations on turbidity ($P<0.05$). In color, the meaningful combination is speed and time. Meanwhile, COD did not show a significant effect on all treatment combinations. These results indicate that the dose plays a major role in the coagulation-flocculation process. Figure 2 shows that the flocculation achieved was highly dose-dependent, and the pollutant removal rate increased to the highest concentration (300 mg/L).

The primary flocculation mechanism by polymers is bridging, which occurs by the adsorption of polymer molecules at more than one site on a particle or sites on different particles. The chain of the polymer must be long, to extend from one surface of the particle to

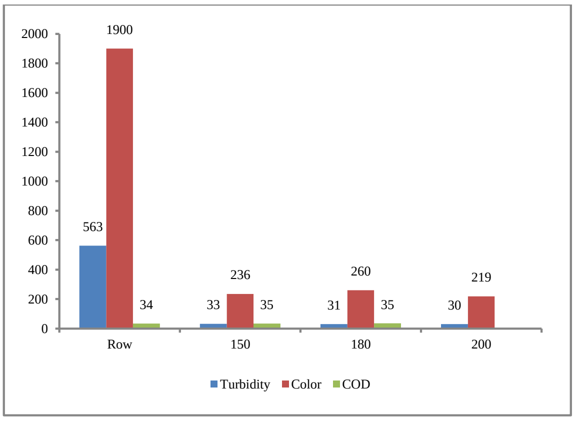
another. According to other studies, the longer the copolymer molecular chain, the more effective it will be [66,70]. Table 2 shows the long chains of natural coagulants.

Table 4. Statistical analysis

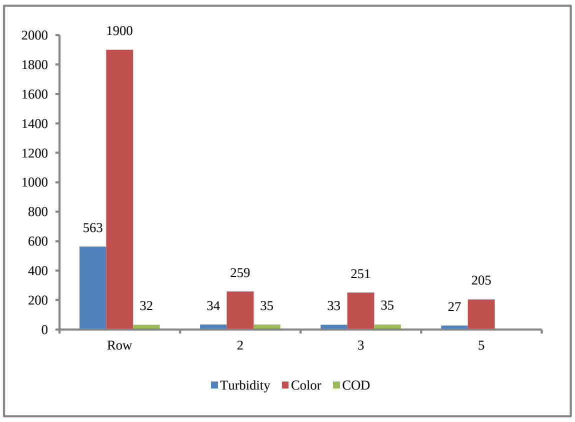
Source	Turbidity		COD		Color	
	Sum of Squares	Sig.	Sum of Squares	Sig.	Sum of Squares	Sig.
Corrected Model	6867.81	0.000	435,93	0,844	357822.18	0.000
Intercept	142380.44	0.000	174842,45	0,000	8178170.06	0.000
Dose	1138.25	0.000	33,32	0,578	26915.74	0.004
Speed	217.60	0.007	36,04	0,346	40823.04	0.000
Time	1252.53	0.000	0,73	0,979	80201.79	0.000
Dose * Speed	404.21	0.005	72,61	0,634	11313.56	0.440
Dose * Time	512.42	0.001	55,36	0,770	22894.81	0.073
Speed * Time	2533.91	0.000	58,23	0,487	154303.16	0.000
Dose * Speed * Time	808.87	0.000	179,62	0,559	21370.05	0.522
Error	2228.32		1815,53		207176.75	



(a)



(b)



(c)

Figure 2. Effect of natural coagulant on the removal turbidity. COD. and color by dosage (a), speed (b), and time (c)

Conclusion

A new copolymer coagulant has been synthesized from banana agricultural waste using microwave radiation. The copolymer performance was characterized by removing turbidity, COD, and color; based on variations in dosage, stirring speed, and stirring time. From the above experimental studies, it can be concluded that natural polysaccharides can be modified by incorporating the cationic moiety on the starch backbone. Furthermore, it becomes an effective flocculation material for treating clean water and wastewater.

Author Contributions

All the authors contributed equally to the preparation, development, and completion of this manuscript.

Ethics

This article is original and contains unpublished material. The corresponding author confirms that the other authors have read and approved the manuscript and that there were no ethical issues involved.

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[OAMJMS] Editor Decision

Assoc. Prof. Dr Sasho Stoleski, MD, PhD via SFS - Journals (Scientific Foundation SPIROSKI - Journals), Skopje, Republic of Macedonia <noreply@publicknowledgeproject.org> 15 Desember 2021 pukul 18.07
Balas Ke: "Assoc. Prof. Dr Sasho Stoleski, MD, PhD" <sstoleski@yahoo.com>
Kepada: Prayudhy Yushananta <prayudhyyushananta@gmail.com>, Mei Ahyanti <mei.ahyanti@gmail.com>

Prayudhy Yushananta, Mei Ahyanti (Author):

We have reached a decision regarding your submission to Open Access Macedonian Journal of Medical Sciences, "Novel Copolymer Cationic from Agroindustrial Waste using Microwave", Manuscript ID = OJS8126.

Our decision is: Revisions Required

Sincerely,
Prof. Dr Mirko Spiroski,
Editor-in-Chief, OAMJMS

Sasho Stoleski

Reviewer A:
Recommendation: Revisions Required

Comments to the Author

The principle of this manuscript is good, but there are improvements in several parts, especially in the results and discussion, and the adjustment of the conclusions to the objectives. thanks

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Reviewer D:
Recommendation: Revisions Required

Comments to the Author

Revision soon

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Novel Copolymer Cationic from Agroindustrial Waste using Microwave

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ABSTRACT

Background. A cationic copolymer has been developed as a substitute for synthetic coagulants, resulting in decreased pH, potential health problems, high costs, and large sludge volumes.
Aim. This study evaluated the potential of banana pith in several treatments as a natural coagulant to reduce turbidity, COD, and color.
Methods. The synthesis was carried out by inserting the cationic moiety of GTA (3-Chloro-2-hydroxypropyl trimethyl ammonium chloride) into the starch backbone by microwave radiation.
Resulth. It has been found that the floculation characteristics depend on the charge neutralization, followed by the linkage between the copolymer chains. The results showed that the initial dose and concentration influenced the copolymer's flocculation performance.
Conclusions. Natural polysaccharides can be modified becomes an effective flocculation material for treating clean water and wastewater.

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Keywords: Coagulation-flocculation, banana pith, turbidity, COD, color

Introduction

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Water supply is intended so that residents can carry out their activities as humans [1,2]. There are two serious problems in the provision of clean water, namely the increasing population and the rapid rate of urbanization [3–10]. It is estimated that the demand for water needs will increase drastically in the world from 9.7 billion in 2020 to 9.7 billion people in 2050 [11]. In the industrial sector, clean water is needed to reach about 20% of the total water available [3]. Problems in the availability aspect are the increase in water pollution from various toxic pollutants as a consequence of the rapidly growing economic development [6,7,9,12–16]. The main problems in industrial waste disposal are hydraulic overload, temperature extremes, amounts of oil and grease, acids or bases, suspended solids, inorganic or organic materials, toxic, and volatile, odor, or corrosive gases [6,7,9,12–16].

These two problems, encourage massive exploitation of groundwater [16,17], resulting in a reduced volume of groundwater storage, land subsidence, negative impacts on water supply, decreased surface water flow and loss of springs, and loss of wetlands thus threatening the sustainability of supply water [18,19]. Globally, it is estimated that there has been a reduction in groundwater during 1900-2008 of 4.500 km^3 or the equivalent of a sea-level rise of 12.6 mm (more than 6% of the total). Since about 1950 the rate of groundwater reduction has increased significantly with the maximum rate occurring during 2000-2008, averaging about 145 km^3/year, or equivalent to 0.40 mm/year of level increase or 13% of the reported rate was 3.1 mm/year during this last period [20] [18] .

Many technologies have been developed to treat water and wastewater. such as precipitation, adsorption, coagulation, flotation, ion exchange, membrane filtration, and biological and electrolytic methods have been used to remove particles from water [21–34]. However, coagulation and flocculation techniques are the most widely applied technology in the world as a vital step in removing colloid particles, natural organic matter, microorganisms, and inorganic ions present in untreated water [16,30,32,35–38].

A coagulant is a very important material in the coagulation-flocculation process which refers to the agglomeration process of colloidal particles with an average size of 5-200 nm and small suspended solids in water, influenced by several factors such as temperature, ionic strength, pH. type and dose of material, coagulant, size and distribution type, concentration and properties of organic matter and colloid particles in suspension [4,9,10,13,16,22,23,25,26,35,37–46].

Metal salts such as ferric chloride ($FeCl_3$), aluminum sulfate ($Al_2 (SO_4) 3$), Pollyalluminium Chloride (PAC), Polyferrous sulfate (PFS), and poly ferric chloride (PFC) have been widely used to treat water and various wastewater [6,13,35,47,48]. Despite providing great performance in water treatment, these coagulants have several disadvantages such as reducing pH to acid, inefficiency in low temperatures, potential causes of health problems (such as Alzheimer's disease, neurotoxic and carcinogenic), relatively high cost of coagulants, and large sludge volume, and some evidence that iron salts and PFS can accelerate pipe corrosion [6,12,14,27,35,44,49,50]. Also, the chlorination method used in large quantities due to the use of high coagulant doses can produce several by-products with long-term harmful effects [10,12].

An alternative is needed to overcome them, one of which is to use plant-based coagulants because they are not toxic or corrosive [7,10,12,45,47,51–54]. Several natural polyelectrolytes have been studied as coagulants or flocculants in water and wastewater treatment with varying results. including rice bagasse [55]. *Tamarindus indica* [56]. *Moringa aloleifera* [57]. *Manihot esculenta* [58]. *Cactus opuntia* [59]. *Dolichos lablab* [60]. The advantages of using organic polyelectrolytes among others, the use of a lower coagulant dose, a smaller increase in ionic load on treated water, a decrease in the aluminum content in treated water, and cost savings [30,61].

Agricultural waste that has not been widely studied as raw material for natural coagulants is banana stem waste. Even though it is very potential because of its abundant and sustainable post-harvest availability [16,30,35,36,62]. The main ingredients in banana pith are water. Fibe, and polysaccharides or starch. The composition of C, H, O as much as 33.2%; 0.49%; and 6.17% [62], other studies mention 32.3% carbon, 4.21% hydrogen, 1.46% nitrogen, 43.5% oxygen, and 0.86% sulfur [30].

Polysaccharides are usually modified by conventional redox grafting methods [22,63–65]. The use of microwave radiation can be used in starch modification based on the advantages of energy saving, and speed [66–68]. In this study, starch from banana pith was synthesized through a copolymerization process using the microwave method, and at the same time, its flocculation performance was studied. The aim of this study was to obtain a natural coagulant that is modified from agricultural waste to reduce turbidity, chemical oxygen demand (COD), and color.

Materials and Methods

This study was conducted using the Way Kuripan River water which is the source of raw water for drinking water treatment (Figure 1). The water sample was dosed with natural coagulant followed by rapid mixing, flocculation, and sedimentation. The efficiency of removing contaminants is calculated by reducing turbidity, chemical oxygen demand (COD), and color. Banana pith is obtained from cavendish *(Musa paradisiaca)* harvested in Central Lampung Regency, Lampung Province.

GTA (3-Chloro-2-hydroxypropyl trimethyl ammonium chloride), sodium hydroxide, and ethanol were obtained from Sigma Aldrich. All these reagents were of analytical grade and

were applied directly. All solutions were prepared by deionized water. An Electrulox (Model No. EMM20M38GW) domestic microwave oven was used for the experiment.

Water sampling

Samples were collected from the Way Kuripan River in pre-cleaned containers (Fig. 1). The samples were stored in a cool box and transported to the laboratory. Sampling using the Standard Method [69].

Figure 1. Sampling coordinates (-5.4373308.105.2515303)

Starch of banana pith

Banana pith cleaned and rinsed with clean water and cut into small pieces. Soak with 0.5% Sodium Metabisulfite ($Na_2S_2O_5$) solution for 15 minutes to stop the browning effect. Maceration (grated). Squeezed, and filtered (0.4 mm) was performed to separate the liquid from the pulp. Do sedation so that the starch settles, the water is removed. Rinse the sediment with distilled water to clean the starch from the fibers. Dry in the oven at 80^0C until dry. Milling and shifting to get banana pith starch flour. Storage of banana pith flour in a desiccator to avoid additional moisture content.

Synthesis of natural coagulant with microwave radiation

Typically. 1.50g GTA (3-Chloro-2-hydroxypropyl trimethyl ammonium chloride) was added to a 100 mL container containing 5 mL sodium hydroxide (0.10 g/mL) aqueous. The solution was stirred thoroughly for 10 min. 10.00 g of starch is added to the mixture. Stirring was continued for 30 minutes on a water bath at 50^0C. Then, microwave irradiation was performed. Microwave irradiation (480 watts) has for 5 minutes at regular intervals. The microwave irradiation cycle is carried out to prevent the temperature from rising above 50^0C. The reaction vessel and its contents were cooled to room temperature. The precipitate is left in the reaction vessel and washed (3 times) with ethanol. Dry in an oven at 50^0C for 6 hours to obtain cationic starch.

Coagulant-flocculation test

Coagulation and flocculation tests were performed on water samples from the Way Kuripan River using the jar test. 500 mL river water was transferred to a 1000 ml beaker glass and placed in a jar-testing device. An initial pH check was performed, and all experiments were carried out at room temperature.

The desired amount of coagulant (Table 1) was added to the sample water and stirred at three variations of speed (rotary per minute) and time (minute). Followed by slow stirring at 30 rpm for 15 min. The mixture was left for 30 minutes to settle. The formation of floc particles was observed and recorded. Samples were taken with a pipette from a depth of 2 cm to determine turbidity, COD, and color. All analyzes were performed in quadruplicate.

Tabel 1. Dosage coagulants tested

Dosage (mg/L)	Speed (rpm)	Time (min)
50	150	2
100	180	3
200	200	5
300		

Analysis water samples

The Mettler Toledo brand pH meter is used to measure the pH value of the sample. The turbidity test used a portable turbidimeter (Model HACH 2100P). The principle of measuring turbidity is the ratio of the light intensity that is scattered by the sample. The sample is placed into the turbidimeter and the turbidity value is shown in NTU units. The total percentage of turbidity removal is calculated:

$$\text{Total turbidity percentage removal} = \frac{A-B}{A} \times 100 \qquad (1)$$

where A is the initial water turbidity (NTU). B is the turbidity after treatment (NTU).

The COD test was measured using UV Spectrophotometer HACH model. Chemical oxygen demand (COD) refers to the amount of oxygen required to oxidize the organic compounds in a water sample to carbondioxide and water. COD percentage removal was calculated as follows:

$$\text{COD percentage removal} = \frac{A-B}{A} \times 100 \qquad (2)$$

where A is the COD of row water (mg/L), B is COD after treatment (mg/L).

Statistical analysis

The effect of dose on turbidity. COD. color was analyzed two-way ANOVA, using software SPSS 24.0. Mathematical model assumption for a design is :

$$Y_{ijk} = \mu + \beta_i + \tau_j + \gamma_k + (\beta\tau)_{ij} + (\beta\gamma)_{ik} + (\tau\gamma)_{jk} + (\beta\tau\gamma)_{ijk} + \varepsilon_{ijk} \qquad (3)$$

i = 1,2,3,4; j = 1,2,3; k = 1,2,3, Y_{ijk}= the response of ith level factor A, jth level factor B, and kth level factor C; μ= Grand mean; β_i = the effect of the ith level factor A; τ_j= the effect of the jth level factor B; γ_k = the effect of the kth level factor B; $(\beta\tau)_{ij}$ = the effect of the combination of ith level factor A with jth level factor B; $(\beta\gamma)_{ik}$ = the effect of the combination of ith level factor A with kth level factor C; $(\tau\gamma)_{jk}$ = the effect of the combination of jth level factor B with kth level factor C; $(\beta\tau\gamma)_{ijk}$ = the effect of the combination of ith level factor A with jth level factor B with kth level factor C; ε_{ij} = Error.

Results and Discussion

Characteric of natural coagulant

The copolymer is synthesized with microwave power (480 watts). The mechanism of microwave-assisted grafting is as follows: The microwave radiation rotates the GTA molecules, leading to the severing of its epoxypropyl bond, and the pi bond electron cloud splits up into two localized clouds (i.e., free radical sites on the constituent carbon atoms) [66,67]. Moreover, the polar groups absorb the microwave (e.g., OH groups attached to the starch molecule). Then behave as if they were anchored to an immobile raft. and its immobile localized rotations will occur in the microwave region [66], which eventually leads to the severing of these bonds, leading to the formation of free radical sites. The free radical sites created on the starch backbone and the GTA interact to yield the cationic graft copolymer through the usual free radical reaction mechanism.

Table 2 shows the high levels of amylopectin in banana pith starch. The grafting of positive ions into amylopectin will produce a more effective polymer because of the dangling polymer chains and highly branched structure [66,70]. The results of the synthesis gave a decrease in amylopectin by 9.87%. Indicates the occurrence of polymer chain termination due to microwave irradiation.

Table 2. Characteristics of banana pith starch

Characteristic	Starch (%)	Amylosa (%)	Amylopectin (%)
Row Starch	47.57	22.95	77.05
Syntesis starch	36.46	30.56	69.44

Treatment of water using natural coagulant

In general, the water quality of the Way Kuripan river shows that it does not meet the quality requirements for drinking water sources (Table 3). Water has significant turbidity, chemical oxygen demand (COD), and color values. Coagulation and flocculation of river water with natural coagulant showed a decrease of 94.4, 19.4, 87.5% of turbidity, COD, and color (Table 4). Even though the quality standards of clean water and drinking water have been achieved, the use of natural coagulants has significantly reduced turbidity, COD, and color.

Table 3. Characteristics of banana pith starch

Parameters	Row water	Treated water	Removal (%)	Guidelines for	
				Clean water	Drinking water
Turbidity (NTU)	563.0	31.4	94.4	25	5
COD (mg/L)	43.2	32.8	19.4	-	-
Color (TCU)	1900.0	238.2	87.5	50	15

Effect of natural coagulant on the removal turbidity. COD. and color

Table 4 shows the significant effect of dose on the decrease in turbidity and color ($p<0.05$), in contrast to the decrease in COD ($P>0.05$). Similarly, the effect of stirring speed and stirring time also significantly affected turbidity and color ($P<0.05$).

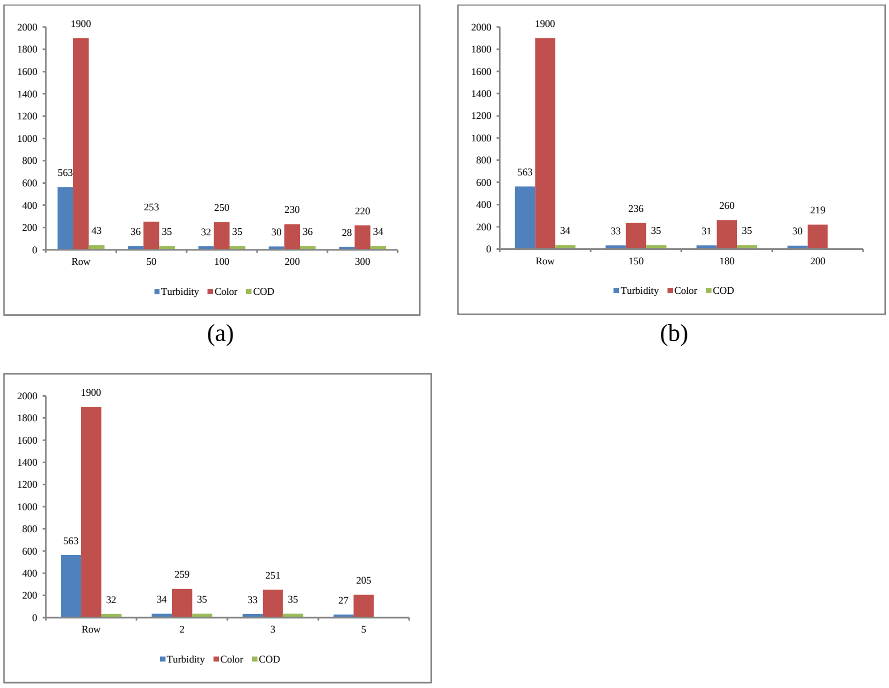
The combination of treatments was also observed in the experiment. A significant effect was found in all treatment combinations on turbidity ($P<0.05$). In color, the meaningful combination is speed and time. Meanwhile, COD did not show a significant effect on all treatment combinations. These results indicate that the dose plays a major role in the coagulation-flocculation process. Figure 2 shows that the flocculation achieved was highly dose-dependent, and the pollutant removal rate increased to the highest concentration (300 mg/L).

The primary flocculation mechanism by polymers is bridging, which occurs by the adsorption of polymer molecules at more than one site on a particle or sites on different particles. The chain of the polymer must be long, to extend from one surface of the particle to

another. According to other studies, the longer the copolymer molecular chain, the more effective it will be [66,70]. Table 2 shows the long chains of natural coagulants.

Table 4. Statistical analysis

Source	Turbidity		COD		Color	
	Sum of Squares	Sig.	Sum of Squares	Sig.	Sum of Squares	Sig.
Corrected Model	6867.81	0.000	435,93	0,844	357822.18	0.000
Intercept	142380.44	0.000	174842,45	0,000	8178170.06	0.000
Dose	1138.25	0.000	33,32	0,578	26915.74	0.004
Speed	217.60	0.007	36,04	0,346	40823.04	0.000
Time	1252.53	0.000	0,73	0,979	80201.79	0.000
Dose * Speed	404.21	0.005	72,61	0,634	11313.56	0.440
Dose * Time	512.42	0.001	55,36	0,770	22894.81	0.073
Speed * Time	2533.91	0.000	58,23	0,487	154303.16	0.000
Dose * Speed * Time	808.87	0.000	179,62	0,559	21370.05	0.522
Error	2228.32		1815,53		207176.75	



(a) (b)

(c)

Figure 2. Effect of natural coagulant on the removal turbidity. COD. and color by dosage (a), speed (b), and time (c)

Conclusion

A new copolymer coagulant has been synthesized from banana agricultural waste using microwave radiation. The copolymer performance was characterized by removing turbidity, COD, and color; based on variations in dosage, stirring speed, and stirring time. From the above experimental studies, it can be concluded that natural polysaccharides can be modified by incorporating the cationic moiety on the starch backbone. Furthermore, it becomes an effective flocculation material for treating clean water and wastewater.

Author Contributions

All the authors contributed equally to the preparation, development, and completion of this manuscript.

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Ethics

This article is original and contains unpublished material. The corresponding author confirms that the other authors have read and approved the manuscript and that there were no ethical issues involved.

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Novel Copolymer Cationic from Agroindustrial Waste using Microwave

ABSTRACT

Background. A cationic copolymer has been developed as a substitute for synthetic coagulants, resulting in decreased pH, potential health problems, high costs, and large sludge volumes.
Aim. This study evaluated the potential of banana pith in several treatments as a natural coagulant to reduce turbidity, COD, and color.
Methods. The synthesis was carried out by inserting the cationic moiety of GTA (3-Chloro-2-hydroxypropyl trimethyl ammonium chloride) into the starch backbone by microwave radiation.
Resulth. It has been found that the floculation characteristics depend on the charge neutralization, followed by the linkage between the copolymer chains. The results showed that the initial dose and concentration influenced the copolymer's flocculation performance.
Conclusions. Natural polysaccharides can be modified becomes an effective flocculation material for treating clean water and wastewater.

Keywords: Coagulation-flocculation, banana pith, turbidity, COD, color

Introduction

Water supply is intended so that residents can carry out their activities as humans [1,2]. There are two serious problems in the provision of clean water, namely the increasing population and the rapid rate of urbanization [3–10]. It is estimated that the demand for water needs will increase drastically in the world from 9.7 billion in 2020 to 9.7 billion people in 2050 [11]. In the industrial sector, clean water is needed to reach about 20% of the total water available [3]. Problems in the availability aspect are the increase in water pollution from various toxic pollutants as a consequence of the rapidly growing economic development [6,7,9,12–16]. The main problems in industrial waste disposal are hydraulic overload, temperature extremes, amounts of oil and grease, acids or bases, suspended solids, inorganic or organic materials, toxic, and volatile, odor, or corrosive gases [6,7,9,12–16].

These two problems, encourage massive exploitation of groundwater [16,17], resulting in a reduced volume of groundwater storage, land subsidence, negative impacts on water supply, decreased surface water flow and loss of springs, and loss of wetlands thus threatening the sustainability of supply water [18,19]. Globally, it is estimated that there has been a reduction in groundwater during 1900-2008 of 4.500 km^3 or the equivalent of a sea-level rise of 12.6 mm (more than 6% of the total). Since about 1950 the rate of groundwater reduction has increased significantly with the maximum rate occurring during 2000-2008, averaging about 145 km^3/year, or equivalent to 0.40 mm/year of level increase or 13% of the reported rate was 3.1 mm/year during this last period [20] [18] .

Many technologies have been developed to treat water and wastewater. such as precipitation, adsorption, coagulation, flotation, ion exchange, membrane filtration, and biological and electrolytic methods have been used to remove particles from water [21–34]. However, coagulation and flocculation techniques are the most widely applied technology in the world as a vital step in removing colloid particles, natural organic matter, microorganisms, and inorganic ions present in untreated water [16,30,32,35–38].

A coagulant is a very important material in the coagulation-flocculation process which refers to the agglomeration process of colloidal particles with an average size of 5-200 nm and small suspended solids in water, influenced by several factors such as temperature, ionic strength, pH. type and dose of material, coagulant, size and distribution type, concentration and properties of organic matter and colloid particles in suspension [4,9,10,13,16,22,23,25,26,35,37–46].

Metal salts such as ferric chloride ($FeCl_3$), aluminum sulfate (Al_2 (SO_4) 3), Pollyalluminium Chloride (PAC), Polyferrous sulfate (PFS), and poly ferric chloride (PFC) have been widely used to treat water and various wastewater [6,13,35,47,48]. Despite providing great performance in water treatment, these coagulants have several disadvantages such as reducing pH to acid, inefficiency in low temperatures, potential causes of health problems (such as Alzheimer's disease, neurotoxic and carcinogenic), relatively high cost of coagulants, and large sludge volume, and some evidence that iron salts and PFS can accelerate pipe corrosion [6,12,14,27,35,44,49,50]. Also, the chlorination method used in large quantities due to the use of high coagulant doses can produce several by-products with long-term harmful effects [10,12].

An alternative is needed to overcome them, one of which is to use plant-based coagulants because they are not toxic or corrosive [7,10,12,45,47,51–54]. Several natural polyelectrolytes have been studied as coagulants or flocculants in water and wastewater treatment with varying results. including rice bagasse [55]. *Tamarindus indica* [56]. *Moringa aloleifera* [57]. *Manihot esculenta* [58]. *Cactus opuntia* [59]. *Dolichos lablab* [60]. The advantages of using organic polyelectrolytes among others, the use of a lower coagulant dose, a smaller increase in ionic load on treated water, a decrease in the aluminum content in treated water, and cost savings [30,61].

Agricultural waste that has not been widely studied as raw material for natural coagulants is banana stem waste. Even though it is very potential because of its abundant and sustainable post-harvest availability [16,30,35,36,62]. The main ingredients in banana pith are water. Fibe, and polysaccharides or starch. The composition of C, H, O as much as 33.2%; 0.49%; and 6.17% [62], other studies mention 32.3% carbon, 4.21% hydrogen, 1.46% nitrogen, 43.5% oxygen, and 0.86% sulfur [30].

Polysaccharides are usually modified by conventional redox grafting methods [22,63–65]. The use of microwave radiation can be used in starch modification based on the advantages of energy saving, and speed [66–68]. In this study, starch from banana pith was synthesized through a copolymerization process using the microwave method, and at the same time, its flocculation performance was studied. The aim of this study was to obtain a natural coagulant that is modified from agricultural waste to reduce turbidity, chemical oxygen demand (COD), and color.

Materials and Methods

This study was conducted using the Way Kuripan River water which is the source of raw water for drinking water treatment (Figure 1). The water sample was dosed with natural coagulant followed by rapid mixing, flocculation, and sedimentation. The efficiency of removing contaminants is calculated by reducing turbidity, chemical oxygen demand (COD), and color. Banana pith is obtained from cavendish *(Musa paradisiaca)* harvested in Central Lampung Regency, Lampung Province.

GTA (3-Chloro-2-hydroxypropyl trimethyl ammonium chloride), sodium hydroxide, and ethanol were obtained from Sigma Aldrich. All these reagents were of analytical grade and

were applied directly. All solutions were prepared by deionized water. An Electrulox (Model No. EMM20M38GW) domestic microwave oven was used for the experiment.

Water sampling

Samples were collected from the Way Kuripan River in pre-cleaned containers (Fig. 1). The samples were stored in a cool box and transported to the laboratory. Sampling using the Standard Method [69].

Figure 1. Sampling coordinates (-5.4373308.105.2515303)

Starch of banana pith

Banana pith cleaned and rinsed with clean water and cut into small pieces. Soak with 0.5% Sodium Metabisulfite ($Na_2S_2O_5$) solution for 15 minutes to stop the browning effect. Maceration (grated). Squeezed, and filtered (0.4 mm) was performed to separate the liquid from the pulp. Do sedation so that the starch settles, the water is removed. Rinse the sediment with distilled water to clean the starch from the fibers. Dry in the oven at 80^0C until dry. Milling and shifting to get banana pith starch flour. Storage of banana pith flour in a desiccator to avoid additional moisture content.

Synthesis of natural coagulant with microwave radiation

Typically. 1.50g GTA (3-Chloro-2-hydroxypropyl trimethyl ammonium chloride) was added to a 100 mL container containing 5 mL sodium hydroxide (0.10 g/mL) aqueous. The solution was stirred thoroughly for 10 min. 10.00 g of starch is added to the mixture. Stirring was continued for 30 minutes on a water bath at 50^0C. Then, microwave irradiation was performed. Microwave irradiation (480 watts) has for 5 minutes at regular intervals. The microwave irradiation cycle is carried out to prevent the temperature from rising above 50^0C. The reaction vessel and its contents were cooled to room temperature. The precipitate is left in the reaction vessel and washed (3 times) with ethanol. Dry in an oven at 50^0C for 6 hours to obtain cationic starch.

Coagulant-flocculation test

Coagulation and flocculation tests were performed on water samples from the Way Kuripan River using the jar test. 500 mL river water was transferred to a 1000 ml beaker glass and placed in a jar-testing device. An initial pH check was performed, and all experiments were carried out at room temperature.

The desired amount of coagulant (Table 1) was added to the sample water and stirred at three variations of speed (rotary per minute) and time (minute). Followed by slow stirring at 30 rpm for 15 min. The mixture was left for 30 minutes to settle. The formation of floc particles was observed and recorded. Samples were taken with a pipette from a depth of 2 cm to determine turbidity, COD, and color. All analyzes were performed in quadruplicate.

Tabel 1. Dosage coagulants tested

Dosage (mg/L)	Speed (rpm)	Time (min)
50	150	2
100	180	3
200	200	5
300		

Analysis water samples

The Mettler Toledo brand pH meter is used to measure the pH value of the sample. The turbidity test used a portable turbidimeter (Model HACH 2100P). The principle of measuring turbidity is the ratio of the light intensity that is scattered by the sample. The sample is placed into the turbidimeter and the turbidity value is shown in NTU units. The total percentage of turbidity removal is calculated:

$$\text{Total turbidity percentage removal} = \frac{A-B}{A} \times 100 \qquad (1)$$

where A is the initial water turbidity (NTU). B is the turbidity after treatment (NTU).

The COD test was measured using UV Spectrophotometer HACH model. Chemical oxygen demand (COD) refers to the amount of oxygen required to oxidize the organic compounds in a water sample to carbondioxide and water. COD percentage removal was calculated as follows:

$$\text{COD percentage removal} = \frac{A-B}{A} \times 100 \qquad (2)$$

where A is the COD of row water (mg/L), B is COD after treatment (mg/L).

Statistical analysis

The effect of dose on turbidity. COD. color was analyzed two-way ANOVA, using software SPSS 24.0. Mathematical model assumption for a design is :

Commented [A1]: This formula has not appeared in the results and discussion

$$Y_{ijk} = \mu + \beta_i + \tau_j + \gamma_k + (\beta\tau)_{ij} + (\beta\gamma)_{ik} + (\tau\gamma)_{jk} + (\beta\tau\gamma)_{ijk} + \varepsilon_{ijk} \qquad (3)$$

i = 1,2,3,4; j = 1,2,3; k = 1,2,3, Y_{ijk}= the response of i^{th} level factor A, j^{th} level factor B, and k^{th} level factor C; μ= Grand mean; β_i = the effect of the i^{th} level factor A; τ_j= the effect of the j^{th} level factor B; γ_k = the effect of the k^{th} level factor B; $(\beta\tau)_{ij}$ = the effect of the combination of i^{th} level factor A with j^{th} level factor B; $(\beta\gamma)_{ik}$ = the effect of the combination of i^{th} level factor A with k^{th} level factor C; $(\tau\gamma)_{jk}$ = the effect of the combination of j^{th} level factor B with k^{th} level factor C; $(\beta\tau\gamma)_{ijk}$ = the effect of the combination of i^{th} level factor A with j^{th} level factor B with k^{th} level factor C; ε_{ij} = Error.

Results and Discussion

Characteric of natural coagulant

The copolymer is synthesized with microwave power (480 watts). The mechanism of microwave-assisted grafting is as follows: The microwave radiation rotates the GTA

Commented [A2]: this should have been explained in the method molecules, leading to the severing of its epoxypropyl bond, and the pi bond electron cloud splits up into two localized clouds (i.e., free radical sites on the constituent carbon atoms) [66,67]. Moreover, the polar groups absorb the microwave (e.g., OH groups attached to the starch molecule). Then behave as if they were anchored to an immobile raft. and its immobile localized rotations will occur in the microwave region [66], which eventually leads to the severing of these bonds, leading to the formation of free radical sites. The free radical sites created on the starch backbone and the GTA interact to yield the cationic graft copolymer through the usual free radical reaction mechanism.

Table 2 shows the high levels of amylopectin in banana pith starch. The grafting of positive ions into amylopectin will produce a more effective polymer because of the dangling polymer chains and highly branched structure [66,70]. The results of the synthesis gave a decrease in amylopectin by 9.87%. Indicates the occurrence of polymer chain termination due to microwave irradiation.

Table 2. Characteristics of banana pith starch

Characteristic	Starch (%)	Amylosa (%)	Amylopectin (%)
Row Starch	47.57	22.95	77.05
Syntesis starch	36.46	30.56	69.44

Treatment of water using natural coagulant

In general, the water quality of the Way Kuripan river shows that it does not meet the quality requirements for drinking water sources (Table 3). Water has significant turbidity, chemical oxygen demand (COD), and color values. Coagulation and flocculation of river water with natural coagulant showed a decrease of 94.4, 19.4, 87.5% of turbidity, COD, and color (Table 4). Even though the quality standards of clean water and drinking water have been achieved, the use of natural coagulants has significantly reduced turbidity, COD, and color.

Table 3. Characteristics of banana pith starch

Parameters	Row water	Treated water	Removal (%)	Guidelines for	
				Clean water	Drinking water
Turbidity (NTU)	563.0	31.4	94.4	25	5
COD (mg/L)	43.2	32.8	19.4	-	-
Color (TCU)	1900.0	238.2	87.5	50	15

Effect of natural coagulant on the removal turbidity. COD. and color

Table 4 shows the significant effect of dose on the decrease in turbidity and color ($p<0.05$), in contrast to the decrease in COD ($P>0.05$). Similarly, the effect of stirring speed and stirring time also significantly affected turbidity and color ($P<0.05$).

The combination of treatments was also observed in the experiment. A significant effect was found in all treatment combinations on turbidity ($P<0.05$). In color, the meaningful combination is speed and time. Meanwhile, COD did not show a significant effect on all treatment combinations. These results indicate that the dose plays a major role in the coagulation-flocculation process. Figure 2 shows that the flocculation achieved was highly dose-dependent, and the pollutant removal rate increased to the highest concentration (300 mg/L).

The primary flocculation mechanism by polymers is bridging, which occurs by the adsorption of polymer molecules at more than one site on a particle or sites on different particles. The chain of the polymer must be long, to extend from one surface of the particle to

Commented [A3]: the discussion is still unclear, for example compared to other studies or water quality standards ... so it needs to be added related to the discussion

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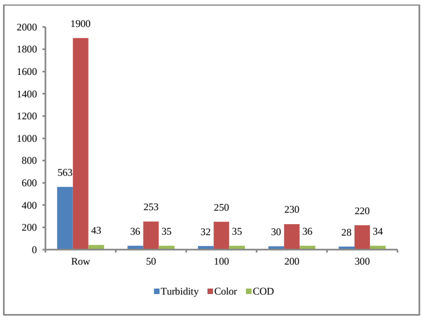
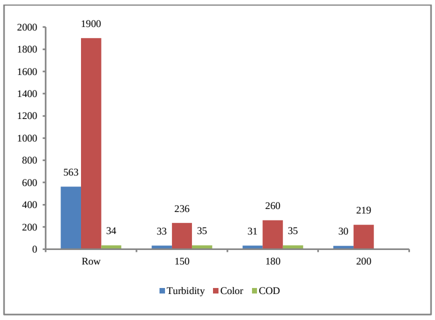
another. According to other studies, the longer the copolymer molecular chain, the more effective it will be [66,70]. Table 2 shows the long chains of natural coagulants.

Table 4. Statistical analysis

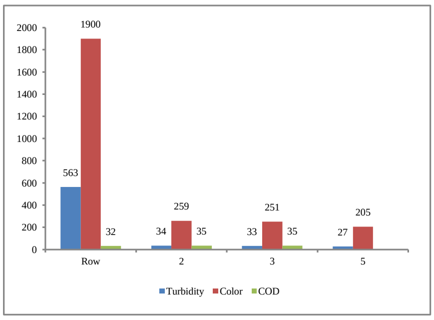
Source	Turbidity Sum of Squares	Turbidity Sig.	COD Sum of Squares	COD Sig.	Color Sum of Squares	Color Sig.
Corrected Model	6867.81	0.000	435,93	0,844	357822.18	0.000
Intercept	142380.44	0.000	174842,45	0,000	8178170.06	0.000
Dose	1138.25	0.000	33,32	0,578	26915.74	0.004
Speed	217.60	0.007	36,04	0,346	40823.04	0.000
Time	1252.53	0.000	0,73	0,979	80201.79	0.000
Dose * Speed	404.21	0.005	72,61	0,634	11313.56	0.440
Dose * Time	512.42	0.001	55,36	0,770	22894.81	0.073
Speed * Time	2533.91	0.000	58,23	0,487	154303.16	0.000
Dose * Speed * Time	808.87	0.000	179,62	0,559	21370.05	0.522
Error	2228.32		1815,53		207176.75	



(a)



(b)



(c)

Figure 2. Effect of natural coagulant on the removal turbidity. COD. and color by dosage (a), speed (b), and time (c)

Conclusion

A new copolymer coagulant has been synthesized from banana agricultural waste using microwave radiation. The copolymer performance was characterized by removing turbidity, COD, and color; based on variations in dosage, stirring speed, and stirring time. From the above experimental studies, it can be concluded that natural polysaccharides can be modified by incorporating the cationic moiety on the starch backbone. Furthermore, it becomes an effective flocculation material for treating clean water and wastewater.

Commented [A5]: he conclusion must answer the research objectives .... it should be written first if the purpose is answered or not, then the description

Author Contributions

All the authors contributed equally to the preparation, development, and completion of this manuscript.

Ethics

This article is original and contains unpublished material. The corresponding author confirms that the other authors have read and approved the manuscript and that there were no ethical issues involved.

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Usability Prototype Smart Portable Nebulizer for Self-care with Respiratory Disorders at Home

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Novel Copolymer Cationic from Agroindustrial Waste using Microwave

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Abstract

BACKGROUND: A cationic copolymer has been developed to substitute synthetic coagulants, resulting in decreased pH, potential health problems, high costs, and large sludge volumes.

AIM: This study evaluated the potential of banana pith in several treatments as a natural coagulant to reduce turbidity, COD, and color.

METHODS: The synthesis was carried out by inserting the cationic moiety of GTA into the starch backbone by microwave radiation. The cationic performance was tested four times, with a factorial design based on dose (4 levels), stirring speed (3 levels), and stirring time (3 levels). A two-way ANOVA test was applied to determine the effect of treatment.

RESULTS: The results showed that the cationic polymer could reduce turbidity (94.4%), COD (19.4%), and color (87.5%). Although all variables showed a significant effect ($p < 0.05$), the use of 300 mg/L dose, stirring speed (200 rpm), for 5 min showed the highest reduction effect.

CONCLUSIONS: Using GTA and microwaves, banana pith starch can be modified into an effective cationic copolymer for water and wastewater treatment.

Introduction

Water supply is intended to carry out their activities as humans [1]. Two serious problems increase the need for clean water, namely, population growth and rapid industrialization growth [2], [3], [4], [5], [6], [7], [8]. It is estimated that global water demand will increase to meet the needs of 9.7 billion people by 2050, from 7.7 billion in 2020 [9]. In the industrial sector, clean water is needed to reach about 22% of the total water available [10]. On the water supply aspect, the problems are the increase in water pollution from various toxic pollutants from industrial and domestic [4], [5], [7], [11], [12], [13], [14], [15]. Domestic wastewater mainly contains pathogenic microbiological, such as Coliform, *E. coli, Streptococcus sp., Pseudomonas sp., Vibrio sp., Clostridia sp., Arcobacter sp., Thiobacillus sp.* [16], [17]. Meanwhile, the industrial wastewater is hydraulic overload, extreme temperatures, oil and grease, acids or bases, suspended solids, inorganic or organic, toxic and volatile materials, odors, or corrosive gases [4], [5], [7], [11], [12], [13], [14], [15].

These two problems encourage massive groundwater exploitation [15], [18]. Resulting in a reduced volume of groundwater storage, land subsidence, negative impacts on water supply, decreased surface water flow and loss of springs, and loss of wetlands, thus threatening to threaten the sustainability of water supply [19], [20]. Globally, it estimated a reduction in groundwater from 1900 to 2008 of 4.500 km^3 (the equivalent of a sea-level rise of 12.6 mm). Since about 1950, the rate of groundwater reduction has increased significantly, averaging about 145 km^3/year (equivalent to 0.40 mm/year of level increase) [19], [21].

Water treatment is the most rational choice to solve the demand and supply of clean water problems, in addition to efforts to diversify sustainable water sources (including the use of rainwater and seawater desalination) [1], [22]. Many technologies have been developed to treat water and wastewater, such as precipitation, adsorption, coagulation, flotation, ion exchange, membrane filtration, and biological and electrolytic methods have been used to remove particles from water [23], [24], [25], [26], [27], [28], [29], [30], [31], [32], [33], [34], [35], [36]. However, coagulation and flocculation techniques are the most widely applied technology globally as a vital step in removing colloid particles, natural organic matter, microorganisms, and inorganic ions present in untreated water [15], [32], [34], [37], [38], [39], [40].

A coagulant is an essential material in the coagulation-flocculation process which refers to the agglomeration process of colloidal particles with an average size of 5−200 nm and small suspended solids in water influenced by several factors such as temperature, ionic strength, pH. type and dose of material, coagulant, size and distribution type, concentration and properties of organic matter, and colloid particles in suspension [2], [7], [8], [12], [15], [24], [25], [27], [28], [37], [38], [39], [40], [41], [42], [43], [44], [45], [46], [47], [48].

Metal salts such as ferric chloride ($FeCl_3$), aluminum sulfate ($Al_2(SO_4)_3$), Pollyalluminium Chloride (PAC), Polyferrous sulfate (PFS), and poly ferric chloride (PFC) have been widely used to treat water and various wastewater [4], [12], [37], [49], [50]. Despite providing good performance in water treatment, the coagulants have several disadvantages such as reducing pH to acid, inefficiency in low temperatures, high cost of coagulants, large sludge volume, and accelerated pipe corrosion. In addition, it also potential causes of health problems (Alzheimer's disease, neurotoxic, and carcinogenic) [4], [11],[13], [29], [37], [46], [51], [52].

An alternative is needed to overcome them: to use plant-based coagulants because they are not toxic or corrosive [5], [8], [11], [47], [49], [53], [54], [55], [56]. Several natural polyelectrolytes have been studied as coagulants or flocculants in water and wastewater treatment with varying results, including rice bagasse [57] *Tamarindus indica* [58], *Moringa aloleifera* [59], *Manihot esculenta* [60], *Cactus opuntia* [61], and *Dolichos lablab* [62]. The advantages of using organic polyelectrolytes include using a lower coagulant dose, a smaller increase in ionic load on treated water, a decrease in the aluminum content in treated water, and cost savings [32], [63].

Agricultural waste that has not been widely studied as raw material for natural coagulants is banana stem waste, even though it is very potential because of its abundant and sustainable post-harvest availability [15], [32], [37], [38], [64]. The main ingredients in banana pith are water, fiber, and polysaccharides or starch. The composition of C, H, O as much as 33.2%; 0.49%; and 6.17% [64], other studies mention 32.3% carbon, 4.21% hydrogen, 1.46% nitrogen, 43.5% oxygen, and 0.86% sulfur [32].

Polysaccharides are usually modified by conventional redox grafting methods [24], [65], [66], [67]. Microwave radiation can be used in starch modification based on the advantages of energy-saving and speed [68], [69], [70]. This study synthesized starch from banana pith through a copolymerization process using the microwave method. Furthermore, the flocculation performance was studied. This study aimed to obtain a natural coagulant modified from agricultural waste to reduce turbidity, chemical oxygen demand (COD), and color.

Materials and Methods

This study was conducted using the Way Kuripan River water which is the source of raw water for drinking water treatment (Figure 1). The water sample was dosed with natural coagulant followed by rapid mixing, flocculation, and sedimentation. The efficiency of removing contaminants is calculated by reducing turbidity, COD, and color. Banana pith is obtained from cavendish *(Musa paradisiaca)* harvested in Central Lampung Regency, Lampung Province.

Figure 1: Sampling coordinates (-5.4373308.105.2515303)

GTA (3-Chloro-2-hydroxypropyl trimethyl ammonium chloride), sodium hydroxide, and ethanol were obtained from Sigma-Aldrich. All these reagents were of analytical grade and were applied directly. All solutions were prepared with deionized water. An Electrolux (Model No. EMM20M38GW) domestic microwave oven was used for the experiment.

Water sampling

Samples were collected from the Way Kuripan River in pre-cleaned containers (Figure 1). The samples were stored in a cool box and transported to the laboratory, sampling using the Standard Method [71].

The starch of banana pith

Banana pith cleaned and rinsed with clean water and cut into small pieces. Soak with 0.5% Sodium Metabisulfite ($Na_2S_2O_5$) solution for 15 min to stop the browning effect. Maceration (grated). Squeezed and filtered (0.4 mm) was performed to separate the liquid from the pulp. Do sedation so that the starch settles, the water is removed. Rinse the sediment with distilled water to clean the starch from the fibers. Dry in the oven at 80°C until dry. Milling and shifting to get banana pith starch flour storage of banana pith flour in a desiccator to avoid additional moisture content.

Synthesis of natural coagulant with microwave radiation

Typically, 1.50g GTA (3-Chloro-2-hydroxypropyl trimethyl ammonium chloride) was added to a 100 mL container containing 5 mL sodium hydroxide (0.10 g/mL) aqueous. The solution was stirred thoroughly for 10 min. 10.00 g of starch is added to the mixture. Stirring was continued for 30 min on a water bath at 500°C. Then, microwave irradiation was performed. Microwave irradiation (480 watts) has for 5 min at regular intervals. The microwave irradiation cycle is carried out to prevent the temperature from rising above 50°C. The reaction vessel and its contents were cooled to room temperature. The precipitate is left in the reaction vessel and washed (3 times) with ethanol. Dry in an oven at 50°C for 6 h to obtain cationic starch.

Coagulant-flocculation test

Coagulation and flocculation tests were performed on water samples from the Way Kuripan River using the Jar-Test. 500 mL river water was transferred to a 1000 ml beaker glass and placed in a jar-testing device. An initial pH check was performed, and all experiments were carried out at room temperature.

The desired amount of coagulant (Table 1) was added to the sample water and stirred at three variations of speed (rotary per minute) and time (minute), followed by slow stirring at 30 rpm for 15 min. The mixture was left for 30 min to settle. The formation of flock particles was observed and recorded. Samples were taken with a pipette from a depth of 2 cm to determine turbidity, COD, and color. All analyses were performed in quadruplicate.

Table 1: Dose of coagulant, speed, and time tested

Dose (mg/L)	Speed (rpm)	Time (min)
50	150	2
100	180	3
200	200	5
300		

Analysis water samples

The turbidity test used a portable turbidimeter (Model HACH 2100P). The principle of measuring turbidity is the ratio of the light intensity scattered by the sample. The sample is placed into the turbidimeter, and the turbidity value is shown in NTU units. The total percentage of turbidity removal is calculated:

$$\text{Total turbidity percentage removal} = \frac{A-B}{A} \times 100 \quad (1)$$

where A is the initial water turbidity (NTU). B is the turbidity after treatment (NTU).

The color test used a colorimeter (model HANNA HI96727). The principle of measuring color is that the amount of light absorbed by a solution is directly proportional to the concentration and width of the solution through which the light passes. The degree of color is determined colorimetrically, with units of TCU (True Color Unit). The total percentage of color removal is calculated:

$$\text{Total color percentage removal} = \frac{A-B}{A} \times 100 \quad (2)$$

where A is the initial water color (TCU). B is the color after treatment (TCU).

The COD test was measured using UV Spectrophotometer HACH model. COD refers to the amount of oxygen required to oxidize the organic compounds in a water sample to carbon dioxide and water. COD percentage removal was calculated as follows:

$$\text{COD percentage removal} = \frac{A-B}{A} \times 100 \quad (3)$$

where A is the COD of row water (mg/L), B is COD after treatment (mg/L).

Statistical analysis

Data were analyzed to determine the individual and combination effects of the research variables (dose, speed, and time) used a two-way analysis of variance (ANOVA) overall analysis at 95% confidence level.

Results and Discussion

Characteristic of natural coagulant

The copolymer is synthesized with microwave power (480 watts). The mechanism of microwave-assisted grafting is as follows: The microwave radiation rotates the GTA molecules, leading to the severing of its epoxy propyl bond, and the pi bond electron cloud splits up into two localized clouds (i.e., free radical sites on the constituent carbon atoms) [68], [69]. Moreover, the polar groups absorb the microwave (e.g., OH groups attached to the starch molecule). Then behave as if they were anchored to an immobile raft. Its immobile localized rotations will occur in the microwave region [68], which eventually leads to the severing of these bonds, forming free radical sites. The free radical sites created on the starch backbone and the GTA interact to yield the cationic graft copolymer through the usual free radical reaction mechanism.

Table 2 shows the high levels of amylopectin in banana pith starch. The grafting of positive ions into amylopectin will produce a more effective polymer because of the dangling polymer chains and highly branched structure [68], [72]. The results of the synthesis gave a decrease in amylopectin by 9.87%. It indicates the occurrence of polymer chain termination due to microwave irradiation.

Table 2: Characteristics of banana pith starch

Characteristic	Starch (%)	Amylosa (%)	Amylopectin (%)
Row starch	47.57	22.95	77.05
Syntesis starch	36.46	30.56	69.44

Treatment of water using natural coagulant

In general, the water quality of the Way Kuripan river shows that it does not meet the quality requirements for drinking water sources (Table 3). Water has significant turbidity, COD, and color values.

Coagulation and flocculation of river water with natural coagulant showed a decrease of 94.4, 19.4, and 87.5% of turbidity, COD, and color (Table 4). Even though the quality standards of clean water and drinking water have been achieved, the use of natural coagulants has significantly reduced turbidity, COD, and color.

Effect of natural coagulant on the removal turbidity, COD, and color

Table 4 shows the significant effect of dose on the decrease in turbidity and color ($p < 0.05$). Similarly, the effect of stirring speed and stirring time also significantly affected turbidity and color ($p < 0.05$). The combination of treatments was also observed in the experiment. A significant effect was found in all treatment combinations on turbidity ($p < 0.05$). In color, the significant effect is only shown in the combination of speed and time.

Table 3: Water quality (pre and post) treatment using natural coagulant

Quality parameters	Row water	Treated water	Removal (%)	Guidelines for	
				Clean water	Drinking water
Turbidity (NTU)	563.0	31.4	94.4	25	5
COD (mg/L)	43.2	32.8	19.4	-	-
Color (TCU)	1900.0	238.2	87.5	50	15

COD: Chemical oxygen demand.

These results indicate that the dose and time of stirring are essential in the flocculation process.

The main mechanisms of the flocculation process of plant-based coagulants are neutralization and bridging [37], [73]. This mechanism begins with the adsorption process of polymer coagulant particles [15], [28], [34], [39], [55]. Bridging usually occurs when long-chain polymers are adsorbed on the particles and leave the coagulant polymer segments to bridge the particles. The high molecular weight is advantageous for the bridging mechanism. The bridging mechanism forms strong clumps, even at elevated agitation. However, irreversible flock damage can occur due to polymer friction under turbulent conditions. Dose plays a vital role in the bridging mechanism. However, at higher doses, the colloidal particles are stabilized due to the steric repulsion of the polymer covering the particles. Meanwhile, there is not enough polymer chain at low doses to form a bridge [37], [40], [61], [73], [74], [75].

Neutralization occurs when the oppositely charged coagulant polymer is adsorbed on the particle surface, thereby neutralizing the charge of the colloidal particles. A high charge density coefficient with low molecular weight will effectively coagulation performance. The reduction in the surface charge of the particles results in a decrease in the electrostatic repulsion between the colloidal particles, thereby enabling coagulation [37], [40], [53], [76].

The raw water quality becomes very important to determine the coagulant dose required [68], [72]. The higher the colloid content, the higher the dose required. However, on the other hand, flocculation cannot exceed the optimum dose, and subsequent additions result in lower efficiency. It is explained that there are no more free particle surfaces available for installation. Hence, it takes the correct dose in water treatment using the flocculation method [28], [68], [77].

Although the removal effect was lower than similar studies, it was due to the different raw water quality. Alwi *et al*. [64] used a 33.3% dose to reduce 98.5% turbidity (81.3 to 1.2 NTU), and 80.1% COD (152.4 to 30.3 mg/L). Kakoi *et al*. [32] used a dose of 0.1 kg/M3 to reduce 98.6% turbidity (279.0 to 4.0 NTU), and 73.0% COD (160.0 to 73.0 mg/L). Villabona *et al*. [78] reduced turbidity by 76.5% (32.3 to 7.6 NTU) at a dose of 33.3%. Meanwhile, the turbidity of the raw water in the study was 563.0 NTU. The high turbidity of the raw water results in a lower removal effect. This factor also occurs in COD.

Figure 2 shows the change in the value of each parameter based on dose, stirring speed, and stirring time. In various doses, the highest reduction effect was at a dose of 300 mg/L, and the lowest was at 50 mg/L. Stirring speed gives the most significant effect at 200 rpm and the smallest at 150 rpm. The stirring time gives the best-removing effect of 5 min. These results explain that the higher the dose used, the higher the reduction effect. It is related to the number of cations available for the bridging mechanism to occur correctly. Stirring speed aims to accelerate the reaction between coagulant and colloid in water. The stirring time is related to the length of flocculation through the bridging mechanism.

However, coagulation/flocculation is a unique process, and the three variables are interrelated. Excessive doses cause stabilization of colloidal particles resulting in steric repulsion from the polymer covering the particles. On the other hand, if the dose is low, there is not enough polymer chain to form a bridge. High speed for a long time causes turbulence, resulting in the rupture of the flock formed. For this reason, Jar

Table 4: Statistical analysis (two-ways analysis of variance)

Source	Turbidity		COD		Color	
	Sum of squares	Significant	Sum of squares	Significant	Sum of squares	Significant
Corrected model	6867.81	0.000	435.93	0.844	357822.18	0.000
Intercept	142380.44	0.000	174842.45	0.000	8178170.06	0.000
Dose	1138.25	0.000	33.32	0.578	26915.74	0.004
Speed	217.60	0.007	36.04	0.346	40823.04	0.000
Time	1252.53	0.000	0.73	0.979	80201.79	0.000
Dose × speed	404.21	0.005	72.61	0.634	11313.56	0.440
Dose × time	512.42	0.001	55.36	0.770	22894.81	0.073
Speed × time	2533.91	0.000	58.23	0.487	154303.16	0.000
Dose × speed × time	808.87	0.000	179.62	0.559	21370.05	0.522
Error	2228.32		1815,53		207176.75	

COD: Chemical oxygen demand.

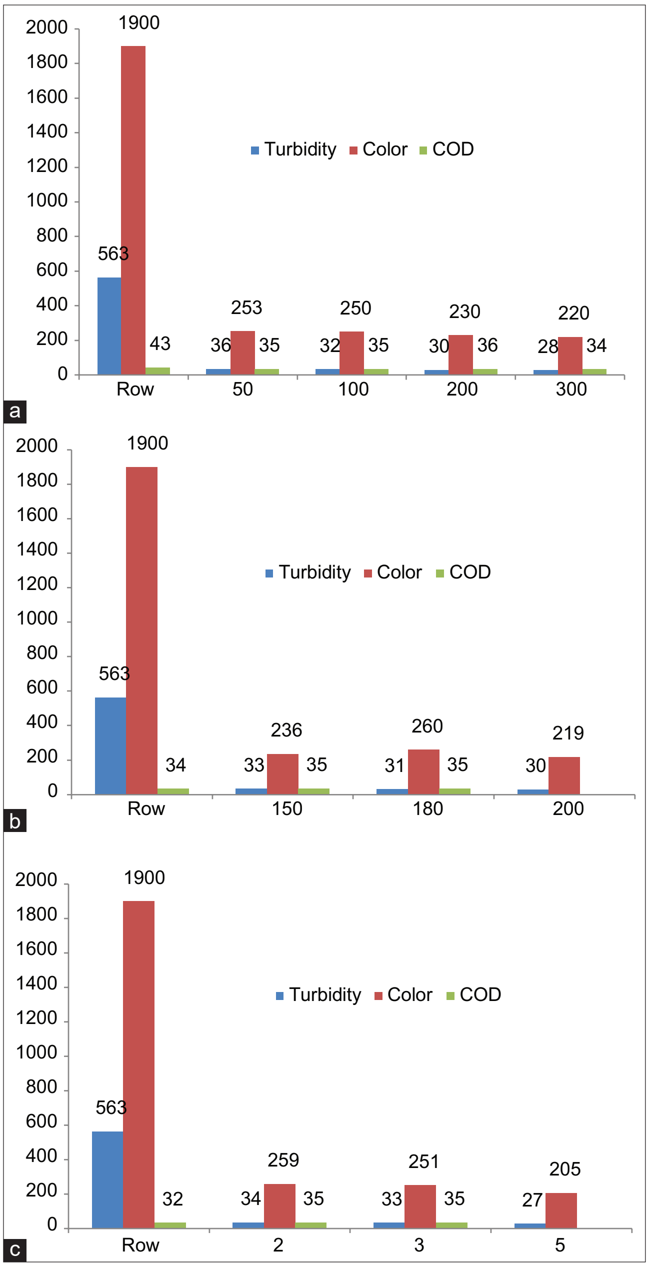

Figure 2: Effect of natural coagulant on the removal turbidity, COD, and color by dose (a), speed (b), and time (c)

Test is needed to get the optimum dose, speed, and time. In this study, the highest reducing effect was at a dose of 300 mg/L, stirring speed of 200 rpm, for 5 min.

Conclusion

A new copolymer coagulant has been synthesized from banana agricultural waste using microwave radiation. The performance of the copolymer was able to remove turbidity (94.4%), COD (19.4%), and color (87.5%). The highest reducing effect was at a dose of 300 mg/L, stirring speed of 200 rpm, for 5 min. This experimental study has proven that natural polysaccharides can be modified by grafting cations into the backbone to be used as effective coagulants for treating water.

Author Contributions

All the authors contributed equally to the preparation, development, and completion of this manuscript.

Ethics

This article is original and contains unpublished material. The corresponding author confirms that the other authors have read and approved the manuscript and that there were no ethical issues involved.

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