Utilization of Banana Pith Starch From Agricultural Waste As A Cationic Coagulant

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Utilization of Banana Pith Starch From Agricultural Waste As A Cationic Coagulant

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The coagulation method is the most commonly used in water treatment. However, long-term use of chemical coagulants can increase the risk of Alzheimer's disease and neurotoxicity, in addition to harming organisms, lowering the pH of the water, corrosion of pipes, and the use of high doses of chlorine. The study synthesized banana pith starch from agricultural waste as a cationic coagulant for river water treatment. Banana pith starch was modified by grafting cations from GTA (3-Chloro-2-hydroxypropyl trimethyl ammonium chloride) into the backbone structure of starch using microwave radiation. Performance tests were carried out on variations in dose (4), speed (3), and stirring time (3). Parameters tested were turbidity, TDS, and color, with four replications. The study found that the synthetic cationic coagulant could reduce turbidity up to 94.4%, while the color and TDS were 87.46% and 57.33%, respectively. The variety of treatments seemed to work on all test parameters ($p<0.05$). However, the most effective treatment was at a dose of 300 ppm, stirring speed of 200 rpm, for 5 minutes. Research has succeeded in proving that banana pith starch can be modified into an effective cationic coagulant to remove colloid compounds in water.

Kata kunci:

Empulur pisang
Kationik
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Kekeruhan
Warna
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A B S T R A K

Saat ini metode koagulasi merupakan metode yang paling umum digunakan dalam pengolahan air. Namun, penggunaan koagulan kimia jangka panjang dapat meningkatkan risiko penyakit Alzheimer danneurotoksik, selain juga merugikan organisme, pH air menjadi rendah, korosi pipa, penggunaan clorin dosis tinggi. Penelitian bertujuan memanfaatkan pati empulur pisang dari limbah pertanian, sebagai koagulan kationik untuk pengolahan air sungai. Modifikasi pati empulur pisang dilakukan dengan cara mencangkokkan kation dari GTA (3-Chloro-2-hydroxypropyl trimetil amonium klorida) ke dalam struktur tulang punggung pati, menggunakan radiasi gelombang mikro. Pengujian kinerja dilakukan pada variasi dosis (4), kecepatan (3), dan waktu pengadukan (3). Parameter yang diuji adalah kekeruhan, TDS, dan warna, dengan empat kali ulangan. Penelitian mendapatkan bahwa koagulan kationik hasil sintesis mampu mereduksi kekeruhan hingga 94,4%, sedangkan warna dan TDS sebesar 87,46% dan 57,33%. Ragam perlakuan terlihat bekerja pada semua parameter uji ($p<0,05$). Namun begitu, perlakuan paling efektif pada dosis 300 ppm, kecepatan pengadukan 200 rpm, selama 5 menit. Penelitian telah berhasil membuktikan bahwa pati empulur pisang dapat dimodifikasi menjadi koagulan kationik yang efektif untuk menghilangkan senyawa koloid dalam air.

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INTRODUCTION

Population growth and industrialization will impact increasing the fulfillment of clean water needs (Asrafuzzaman, Fakhruddin, & Hossain, 2011; Bhatnagar & Sillanpää, 2010; Carolin, Kumar, Saravanan, Joshiba, & Naushad, 2017; Choy, Prasad, Wu, Raghunandan, & Ramanan, 2014; Hakizimana et al., 2017; Quince, 2015; Rahmani, 2015; Senthil Kumar et al., 2019; Yushananta, 2021). It is estimated that in 2045 Indonesia needs 11.64 gm3/year of clean water for domestic use (population predictions follow BPS (BPS Indonesia, 2018a), water needs to follow PUPR (PUPR, 2007)). Meanwhile, surface water quality is decreasing due to domestic and industrial pollution. This condition will encourage the exploitation of groundwater (Ahmad & Danish, 2018; Tamaddun, Kalra, & Ahmad, 2018), which results in a decrease in the volume of groundwater storage, a decrease in the ground surface level, loss of springs, and the loss of wetlands (Konikow & Kendy, 2005; Mays, 2013).

Indonesia has abundant water availability, reaching 15,500 m3/capita/year, far above the world average (8,000 m3/capita/year) (Mawardi, 2008). However, its quality decreases due to domestic, industrial waste, and soil erosion (Irwan, Alianto, & Toja, 2017; Kristianto, 2017; Utami, Kumala Putri, & Ekayani, 2017). As a result, people use unhealthy water, making them vulnerable to digestive diseases and heavy metals such as *Cu, Pb, Cd, Zn,* and *Hg*(Kristianto, 2017; Kuta, Emigilati, Hassan, & Ibrahim, 2014; Oludairo & Aiyedun, 2016; Taiwo, Adenike, & Aderonke, 2020; Yushananta & Bakri, 2021). Healthy water must meet the physical, chemical, microbiological, and radioactive quality requirements (Kemenkes, 2017), in addition to the adequacy of 60-100 liters/person/day (PUPR, 2007). Until 2019, access to clean water has only reached 64.54% (BAPENAS, 2019).

Until now, technologies have been developed to treat water, such as precipitation, adsorption, coagulation, ion exchange, membrane filters, biological and electrolytic methods (Abdulfatai, Saka, Afolabi, & Micheal, 2013; Barbosa et al., 2018; Choudhary & Neogi, 2017; Ghimici & Constantin, 2020; Gurpilhares, Cinelli, Simas, Pessoa, & Sette, 2019; Kakoi, Kaluli, Ndiba, & Thiong'o, 2016; Li et al., 2016; Muthuraman & Sasikala, 2014; Radoiu, 2004; Salleh, Mahmoud, Karim, & Idris, 2011; Shen, Gao, Guo, & Yue, 2019; Stavrinou, Aggelopoulos, & Tsakiroglou, 2018; Wu, Liu, Yang, & Li, 2016; Yongabi, 2010). However, coagulation and flocculation are the most widely applied technologies because of their convenience and economy (Ahmad & Danish, 2018; Kakoi et al., 2016; Kristianto, 2017; Maurya & Daverey, 2018; Muthuraman & Sasikala, 2014; Sillanpää, Ncibi, Matilainen, & Vepsäläinen, 2018; Tripathy & De, 2006). The coagulation-flocculation process refers to the agglomeration process of colloidal particles, which requires coagulant as the main ingredient. Currently, the coagulants that are widely used are metal-based, such as ferric chloride ($FeCl_3$), aluminum sulfate ($Al_2(SO_4)_3$), Pollyalluminium Chloride (PAC), Polyferrous sulfate (PFS), and poly ferric chloride (PFC) (Choy et al., 2014; De Carvalho et al., 2015; Kristianto, 2017; Oladoja, 2015; Salehizadeh, Yan, & Farnood, 2018).

Despite providing good performance, chemical coagulants can have adverse effects on organisms, lowering the pH of the water, large sludge residues, pipe corrosion. It is also a potential cause of health problems such as Alzheimer's, neurotoxic, carcinogenic (Choubey, Rajput, & Bapat, 2012; Choudhary & Neogi, 2017; Choy et al., 2014; Kristianto, 2017; Mouhamed Bayane, Yanjun, & Bekhzad, 2020; Taiwo et al., 2020; Walton, 2013; Yavuz, Vaizoğlu, & Güler, 2013). In addition, chlorination, due to high coagulant doses, can produce several by-products with long-term harmful effects (Senthil Kumar et al., 2019; Taiwo et al., 2020). Thus, the use of plant-based coagulants is an alternative because it is non-toxic and corrosive (Asrafuzzaman et al., 2011; Baptista et al., 2017; Ibrahim & Yaser, 2019; Kalia & Sabaa, 2013; Oladoja, 2015; Polaskova, Peer, Cermak, & Ponizil, 2019; Priyatharishini, Mokhtar, & Kristanti, 2019; Senthil Kumar et al., 2019; Taiwo et al., 2020).

Several natural polymers have been investigated as coagulants, including rice bagasse (Guo & Chen, 2017), P. mucilage (A. Mishra & Bajpai, 2005), Moringa aloleifera (Rodiño-Arguello, Feria-Diaz, Paternina-Uribe, & Marrugo-Negrete, 2015), Manihot esculenta (dos Santos et al., 2018), Cactus opuntia (Choudhary, Ray, & Neogi, 2019). One of the agricultural wastes that has not been studied much is the banana pith. The main content of banana pith is water and starch (polysaccharide) with a ratio of 20% and 76%. The composition of C, H, O as much as 33.2%; 0.49%; and 6.17% (Alwi, Idris, Musa, & Ku Hamid, 2013). Polysaccharides are natural polymers consisting of a mixture of amylose and amylopectin. The use of pith from banana harvest waste is very potential because of its abundance and sustainability (Ahmad & Danish, 2018; Alwi et al., 2013; Kakoi et al., 2016; Kristianto, 2017; Maurya & Daverey, 2018). In 2018, the national banana production was 7,264,379 tons/year, and the average banana production in Lampung Province from 2017-2019 was 12.5 million clumps (BPS Indonesia, 2018b).

Several previous studies have shown the potential of the banana pith as a coagulant, both in its fresh form (Alwi et al., 2013), pith flour (Kakoi et al., 2016), and hydrolysis of pith flour with sodium hydroxide (Villabona-Ortíz, Tejada-Tovar, & Ortega Toro, 2019). Several drawbacks have also been pointed out, such as using high doses, thereby increasing the organic matter content in the water. For this reason, efforts are needed to improve the performance of banana pith starch as a coagulant, one of which is to modify it into a cationic polymer. Cationic polymers are polymers with a positive charge (Choy et al., 2014; Kalia & Sabaa, 2013; Katrivesis, Karela, Papadakis, & Paraskeva, 2019; Kristijarti, Suharto, & Marieanna, 2013; Oladoja, 2015; Priyatharishini et al., 2019; Sillanpää et al., 2018; Wan Ahmad, 2016; Yaman, 2018; Yavuz et al., 2013), making them suitable for negatively charged colloids (Abidin, Mohd Shamsudin, Madehi, & Sobri, 2013; Baptista et al., 2017; Bolto & Gregory, 2007; Choy et al., 2014; Priyatharishini et al., 2019; Salehizadeh et al., 2018; Villabona-Ortíz et al., 2019). Through a microwave-assisted hydrolysis process, the research aimed to modify banana pith starch from agricultural waste as a cationic polymer. The grafting of positive ions into amylopectin will result in a more effective polymer due to the long and highly branched polymer chains (Bolto & Gregory, 2007).

METHOD

Banana pith starch

Banana pith is cleaned and washed with clean water. Then it was cut into small pieces and soaked in 0.5% sodium metabisulfite ($Na_2S_2O_5$) solution to stop the browning effect. Grated, squeezed, and filtered to separate the liquid and

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dregs. Perform sedation so that the starch settles and the water is removed. Rinse the sediment (starch) with distilled water (repeatedly) to clean the starch from the fibers. It was dried using an oven at a temperature of 60^0C until dry. Milling and sieving were carried out to obtain banana pith starch flour–storing banana pith flour in a desiccator to avoid adding water content.

Cationic starch synthesis

A total of 1.50g of GTA (3-Chloro-2-hydroxypropyl trimethyl ammonium chloride) was added to a container containing 5 mL of sodium hydroxide (0.10 g/mL), stirred for 10 minutes. 10.00 g of starch is added to the mixture. Stirring was continued for 30 minutes on a water bath. Microwave radiation (480 watts) is carried out for 5 minutes at regular intervals. The microwave irradiation cycle is carried out to prevent temperature rise above 50^0C. The reaction vessel and its contents were cooled to room temperature and washed (3 times) with ethanol. It was dried in an oven at 500C for 6 hours to obtain cationic starch.

Coagulation-flocculation

Coagulation and flocculation tests were carried out on water samples (1000 ml) using a jar test. Coagulant doses (50, 100, 200, 300 ppm) were added to the sample water and stirred at three-speed variations (150, 180, 200 rpm) for three-time variations (2, 3, 5 minutes). Then slow stirring (30 rpm) for 15 minutes. The mixture was left for 30 minutes to settle. Samples were taken with a pipette from a depth of 2 cm to determine turbidity, color, and TDS. The experiment was carried out with four replications.

Analysis

Turbidity was measured with a portable turbidimeter (HACH Model 2100Q), color (HANNA HI96727), and TDS (LUSO). The total percentage of pollutant reduction is calculated by:

$$\text{Total Reduction (\%)} = \frac{A-B}{A} \times 100 \quad (1)$$

where A is the initial parameter value of raw water, namely turbidity (NTU), color (TCU), and TDS (ppm), B is the parameter value after treatment.

The effect of treatment (dose, stirring speed, stirring time) on the parameters of turbidity, color, and TDS were analyzed by two-way ANOVA. SPSS 24.0 software is used in the analysis.

RESULTS AND DISCUSSION

Characteristics of cationic coagulants

Starch from banana pith was synthesized by microwave power (480 watts). The mechanism of microwave-assisted grafting is as follows: Microwave radiation will spin the GTA molecule, causing a break in the epoxypropyl bond, and the electron cloud splits into two local clouds (i.e., free radical sites on the constituent carbon atoms) (Lin et al., 2012; S. Mishra, Mukul Sen, & Jha, 2011). Polar groups will absorb microwaves (OH groups attached to starch molecules), which ultimately leads to breaking these bonds, leading to the formation of free radical sites. The free radical sites created on the starch backbone and GTA interact to produce cationic graft polymers via a free radical reaction mechanism (Lin et al., 2012).

A simple explanation, starch samples cationized with GTA under alkaline conditions underwent a 2-step reaction. The first stage is the deprotonation of the OH group in GTA to form negatively charged oxygen, then releasing Cl atoms to produce carbonium ions (positively charged C atoms). The carbonium atom then bonds with negatively charged oxygen to form the compound 2,3-epoxy propyl trimethyl ammonium chloride. In the next step, the epoxy group in the compound will attack the H atom of the OH group on the C atom of the starch structure to produce negatively charged oxygen. Then form a bond with the carbonium atom in 2,3-epoxy propyl trimethyl ammonium chloride resulting in a cationized starch structure.

Table 1 shows the high amylopectin content in banana pith starch. The grafting of positive ions into the amylopectin group will produce a more effective polymer because it has a long and branched-chain structure (Bolto & Gregory, 2007; Lin et al., 2012). The results showed that the synthesis reduced amylopectin levels by about 9.87%. It is indicated that the polymer chains are broken due to microwave radiation.

Table 1
Characteristics of banana pithstarch

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

Water treatment with cationic coagulant

Examination of the Way Kuripan River water showed the quality that did not meet health requirements, with high turbidity, color, and TDS values (Table 2). The study results using cationic coagulants (coagulation and flocculation processes) were able to reduce the relatively high value of pollutant parameters, namely turbidity by 94.40%, color by 87.46%, and TDS by 57.33% (Table 2). Although not all parameters meet the quality standards of clean water and drinking water, the cationic coagulants have significantly reduced the values of the three parameters.

Table 2
Water quality before and after treatment

Parameters	Row water	Treatment	Reduction (%)	Standard	
				Clean water	Drink water
Turbidity (NTU)	563.0	31.4	94.4	25	5
Color (TCU)	1900.0	238.2	87.5	50	15
TDS (ppm)	207.0	88.3	57.3	1000	500

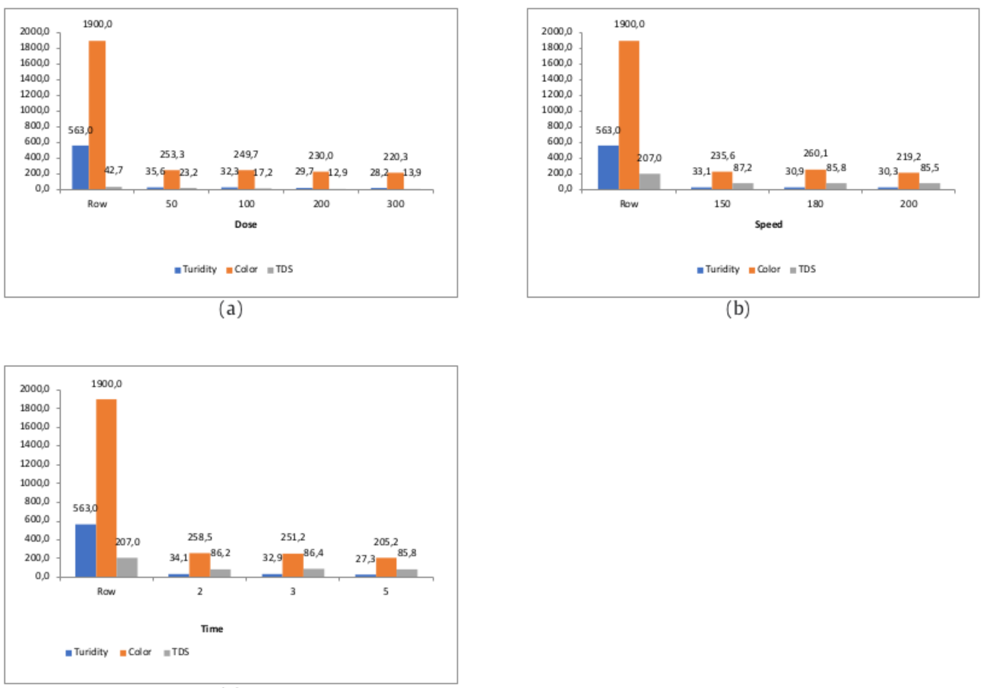
Effect of cationic coagulants on decreasing turbidity, color, and TDS

In Table 3, it can be seen that the dose of coagulant had a significant effect on decreasing the value of turbidity, color, and TDS ($p<0.05$). Similarly, the speed and time of stirring also showed a significant effect ($P<0.05$).

Table 3
Statistically analysis

	Turbidity		Color		TDS	
	Sum of Squares	**Sig.**	**Sum of Squares**	**Sig.**	**Sum of Squares**	**Sig.**
Corrected Model	6867.81	0.000	357822.18	0.000	6843,32	0,000
Intercept	142380.44	0.000	8178170.06	0.000	1123547,00	0,000
Dose	1138.25	0.000	26915.74	0.004	1017,85	0,000
Speed	217.60	0.007	40823.04	0.000	1050,43	0,000
Time	1252.53	0.000	80201.79	0.000	1146,87	0,000

The main mechanism of flocculation by polymers is bridging, which occurs by adsorption of polymer molecules at more than one site on the particle or sites on different particles. The polymer chains must long dangle, bonding from one particle surface to another. The longer the polymer molecular chain, the more effective(Bolto & Gregory, 2007; Lin et al., 2012). Table 1 shows the long chain of cationic coagulants.



(a) (b) (c)

Figure 1. Effect of cationic starch on turbidity, color, and TDS parameters, based on dose (a), stirring speed (b), and time (c).

In Figure 1, it can be seen that the highest reduction was at a dose of 300 ppm (a), stirring speed of 200 rpm (b), for 5 minutes (c). These results explain that the higher the dose used, the more cations contained in the processing so that the bridging mechanism occurs appropriately. Bridging causes the formation of strong bonds between polymers and particles (Almonaityte, Bendoraitiene, Babelyte, Rosliuk, & Rutkaite, 2020; Bolto & Gregory, 2007; Lin et al., 2012; Yongabi, 2010). This study provides better results than previous studies in terms of the use of smaller coagulant doses (Alwi et al., 2013; Bolto & Gregory, 2007; Kakoi et al., 2016; Lin et al., 2012).

On higher doses, the colloidal particles were destabilized due to the steric repulsion of the polymer covering the particles. In contrast, there were not enough polymer chains at lower doses to form bridges. The flocculation also 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. So it takes an adequate dose in water treatment using the flocculation method (Almonaityte et al., 2020; Lin et al., 2012; Yongabi, 2010).

Stirring speed aims to accelerate the reaction between the coagulant and the particles or materials in the water. The positive charge implanted in the polymer will attract colloidal particles, which generally have a negative valence charge. 2. Metal reduction in the research results is due to the presence of metal bonds with particulates so that the reduction of sludge will be followed by metal reduction. However, floc breakdown may occur due to polymer friction under turbulent conditions. Therefore, the proper stirring speed is required.

The stirring time gives the best-removing effect at 5 minutes. This study also confirms from previous studies that stirring for 5 minutes allows the bridging mechanism to work well and produces a strong bond.

LIMITATION OF THE STUDY

This research is the first evidence of the utilization of banana pith waste as a natural coagulant raw material. The effect of pH on treatment, sludge residue characteristics, and coagulation behavior has not been investigated. The whole will be the scope of the subsequent research.

CONCLUSIONS AND SUGGESTIONS

Cationic coagulant has been successfully synthesized by utilizing banana agricultural waste. Cationic coagulant was able to reduce turbidity (94.4%), color (87.5%), and TDS (57.3%). All treatments had a significant effect on parameter values, but the most effective treatment was at a dose of 300 ppm, stirring speed of 180 rpm, for 5 minutes. Research has shown that natural polysaccharides can be modified by grafting cations into the starch backbone using microwaves. The results of the study can be a safe alternative coagulant for water treatment.

ETHICAL CONSIDERATIONS

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Conflict of Interest Statement

The authors declared that they have no conflict of interests.

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