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The Use of Papaya Seeds (Carica papaya L.) as a Biocoagulant in the Dairy Industry Liquid Waste Treatment Process with the Coagulation-Flocculation Method

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Abstract

Dairy industry discharges wastewater characterized by high values of Total Suspended Solid and turbidity. The waste is commonly processed using chemical coagulants which negatively impact human's health and the environment. Papaya seeds can be used as a coagulant as their positively charged proteins that can bind the negatively charged particles allowing the resulting floc to settle and clear water to be obtained. The goals of this study are to determine the characteristics of the dairy industry wastewater before and after flocculation coagulation, the characteristics papaya seed biocoagulant, the effect of the biocoagulant's dosses, particle sizes, and stirring speed on the turbidity and the Total Suspended Solid of the dairy industry wastewater, and the performance of the biocoagulants in processing the dairy industry waste using Sludge Volume Index and Sludge Mass. The treatments applied in this research are in terms of doses (250 mg, 500 mg, and 750 mg), particle sizes (60,120, and 230 mesh), fast stirring speeds (120 rpm, 180rpm, 200 rpm for 1 minute), and slow stirring speeds (10 rpm, 30rpm, and 50 rpm for 20 minutes) all of which were expected to reduce the waste's turbidity and the number of Total Suspended Solid. This study finds that the treatment with the highest reduction of Total Suspended Solid is D3P3KP2, which decreased turbidity from 754 NTU to 92 NTU. The treatment with the highest reduction of Total Suspended Solid was D3P3KP2 which decrease the Total Suspended Solid from 124 mg/L to 79 mg/L. The performance of the papaya seed biocoagulant of D3P3KP2 treatment is considered good as the acquired highest value of the Sludge Volume Index is 83.13 ml/g and as the procured lowest value of the Sludge Mass is 81.48%.

Keywords: biocoagulants, carica papaya, coagulation, flocculation, dairy industry, wastewater

1. Introduction

As dairy industry worldwide is growing tremendously in size and number of factories, it has become one of the largest contributors of wastewater. The waste's concentration varies greatly depending on the production technology, the used equipment, the reusage rate of the wastewater, the loss of raw materials, and the waste management. The amount of the wastewater produced by the industry comes from tank trucks, pipelines, and equipment cleaning at the end of each cycle, the generated wastewater is 70% of the amount of fresh water used in the factory (Slavov et al., 2017). In general, the waste from the diary processing industry contains high concentrations of organic matter such as proteins, carbohydrates, lipids, Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), and Total Suspended Solid (TSS). In addition, the turbidity and the varying pH of the waste require specific treatments in order to prevent or at least to minimize environmental problems (Britz et al., 2006).

The wastewater from dairy industies is treated using physical, chemical and biological methods, and coagulation-flocculation is the most important physicochemical treatment method for reducing colloidal suspended solids and turbidity (Al-Mutairi et al., 2004). In general, the coagulation-flocculation method uses chemical coagulants, namely Iron chloride ($FeCl_3.6H_2O$), Aluminum sulfate ($Al_2[SO_4]3.6H_2O$) and Calcium hydroxide ($Ca[OH]_2$) (Loloei et al., 2014). From a health perspective, due to the non-degradable nature of the chemical coagulants, chemical residues in the treated water can accumulate in bodily cells for a long time and cause health problems such as nervous system failure, Alzheimer and dementia (Ahmad et al., 2016). From an environmental

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perspective, the chemical coagulants are relatively expensive and cause secondary contamination of sludge due to higher levels of chemical residues, and the resulting sludge is toxic (Kakoi et al., 2017).

Alternative coagulants are needed, such as using papaya seeds. The fruit consumption results in the production of food waste as discarded papaya skin and seeds in particular constitutes 15-20% of the fruit (Pesqueira & Nunez, 2017). This kind of waste can be minimized by utilizing it for various purposes including as a biocoagulant in wastewater treatment. Papaya seeds can be used as a coagulant due to their positively charged proteins which can bind negatively charged particles such mud, clay, bacteria, toxins etc, allowing the resulting floc to settle and clear water to be obtained through adsorption and charge neutralization, papaya seed powder has the ability to combine with solids in water and settle to the bottom (Yimer and Bayisa, 2021).

There have been many studies discussing the use of papaya seeds as biocoagulants. Papaya seed as biocoagulant has been applied for purification of river water. About 89.14% removal efficiency of turbidity removal efficiency and total dissolved solids was achieved (George and Chandrn, 2018). Another study of using citrus microcarpa peels and papaya seeds as a natural coagulant has been also reported by Dollah *et al* (2021). The optimum dosage of composite natural coagulant was 90 mg/L using an 80:20 ratio of citrus microcarpa peels and papaya seeds with 97% of turbidity removal.

This study aims to determine the characteristics of dairy industry wastewater, the biocoagulant characteristics of papaya seeds, the effect of biocoagulant dosage, particle size, and stirring speed on turbidity and TSS, and the performance of papaya seed biocoagulants using Sludge Volume Index (SVI) and Sludge Mass (SM). The investigation involved the use of papaya seeds as a biocoagulant for airy industry wastewater treatment with several dosages of coagulant, particle sizes and stirring speeds.

2. Methodology

2.1. Materials and Instruments

The materials used in this study are California papaya seeds obtained from a fruit shop in Malang City and dairy industry wastewater. The instruments used in this study are beaker glass, porcelain cup, jar test JLT6, spatula, Memmert ovens, Whatman papers No.42, 1 Liter bottles, gloves, cool box, blender, Ohaus PAJ 1003 analytical balances, funnels, clamp, sieves with sizes of 60 mesh, 120 mesh, and 230 mesh, and a freezer.

2.2. Sampling and Testing

The wastewater sample used in the study was taken from one of the dairy industries in the city of Malang, namely PT. X. The waste sampling process was carried out by observing the rules for sampling wastewater in accordance with SNI 6989.59:2008. The wastewater sampling method is the grab sampling method or temporary wastewater sampling at one particular location in the canal before entering the wastewater receiving waters. The procured wastewater sample was then taken to the laboratory for further initial testing. It was is stored in a cool box to prevent changes. The parameters for the initial observations were turbidity and TSS test, performed according to SNI 6989.3:2019.

2.3. Preparation and Characteristic Analysis of Biocoagulant

The biocoagulant material used in this study was papaya seeds (Carica papaya L.). To make biocoagulant, the seeds are cleaned by washing, the skin is peeled, then the peeled seeds are dried in an oven at 500C for 24 hours (Amran et al., 2021). The drying process is carried out to obtain a water content below 10% (Amran et al., 2021). The dried seeds are ground into powder and sieved using 60, 120 mesh and 230 mesh sieve. After weighing according to the dosage and the powder is ready to be made.

The biocoagulant was then subjected to characteristic analysis using FTIR (Fourier Transform Infrared Spectroscopy) incorporating Shimadzu IRPrestige21 and Proximate. This analysis helps determine the compounds and the functional groups contained in biocoagulants.

2.4. Coagulation-Flocculation Process

The experiment was carried out by preparing 1000 ml beaker glasses filled with the wastewater. Each beaker glass was given a treatment label containing papaya seed biocoagulant dose, particle size, and mixing speed. In this process, the first this to do was preparing the biocoagulant made from papaya seed in the doses of 250 mg, 500 mg, and 750 mg with particle size of 100, 200, and 250 mesh. Then, the doses were poured to the beaker glasses containing the wastewater sample. The beaker glasses were placed in the Jar Test to be treated with fast mixing are at 120 rpm, 180 rpm and 200 rpm for 1 minute and with slow mixing at 10 rpm, 30 rpm and 50 rpm for 20 minutes. After the mixing was complete, Sludge Volume Index and Sludge Mass measurements were carried out.

2.5. Parameter Analysis

The samples that have undergone the coagulation-flocculation process were tested for turbidity and TSS as the parameters, SVI and SM. Observation of floc deposits that occur using the Sludge Volume Index (SVI) and SM. The TSS analysis was performed using HACH DR900 Colorimeter, and the turbidity analysis was conducted using Turbidimeter TB 300 IR. SVI quantifies the sludge volume after it being deposited for 30 minutes in a 1000 mL Imhoff cone (Zhang et al., 2013). The coagulation and flocculation of the wastewater samples were poured into the Imhoff cone and allowed to stand for 30 minutes. The waste water was is then poured into a Buchner and Whatman paper along with the precipitate. The precipitate that had been filtered on Whatman paper was weighed using an analytical balance to determine the wet weight of the precipitate. The Whatman paper was then put in an oven for 1 hour at 105^0C to obtain its dry residue (Zhang et al., 2013). Sludge Volume Index and SM were used as the determinants of the biocoagulant's performance in wastewater treatment. The SVI was calculated by determining the difference between the masses of wet and dry sludge divided by the acquired mass of the wet sludge (Putra & Ainun, 2021). Turbidity and TSS testing were carried out at the Water Quality and Waste Treatment Laboratory using methods that conforms the SNI 6989.3: 2019.

2.6. Data Analysis

The acquired data was processed to obtain results related to the characteristics of the dairy industry wastewater before and after the treatment, the characteristics of the papaya seed biocoagulant, the effect of increasing the dose, the particle size, and the stirring speed of the biocoagulant on the sample's turbidity and TSS, and the effect of the treatment on the values of SVI and SM. The data regarding the characteristic of the papaya seeds obtained from the results of FTIR and Proximate analysis. The data of the characteristic wastewater was acquired to determine the parameters of its turbidity and TSS before and after the treatment. The analysis on the Sludge Volume Index and SM values was carried out to determine the performance of the papaya seeds biocoagulant against the dairy industry wastewater. The calculation of Sludge Volume Index (SVI) can be seen in Equation 1, while SM is expressed in Equation 2.

$$\mathrm{SVI(ml/g)} = \frac{\mathrm{SV\ (ml/L)x1000}}{\mathrm{TSS\ (mg/L)}} \quad (1)$$

Remarks:
SVI : Sludge Volume Index
SV : Sludge volume
TSS : Total suspended solid

$$\mathrm{SM} = \frac{\mathrm{Sm_0} - \mathrm{Sm_t}}{\mathrm{Sm_0}} \mathrm{x100\ \%} \quad (2)$$

Remarks:
Sm_0 : Wet weight
Sm_1 : Dry weight

3. Results and Discussion

3.1. Characteristics of Papaya Seeds

The analysis of the characteristics of the papaya seeds biocoagulant was carried out using FTIR analysis whose results can be seen in Figure 1. The results of the FTIR analysis on papaya seed powder show a peak at the wave of 1050–1300 cm^{-1} which indicates the presence of C–O Alcohol/Ether/Carboxylic Acid/Ester functional groups which usually appear at that wavelength. The wavelength that appears at 1180–1360 cm^{-1} indicates the presence of C-N Amine/Amide functional group.

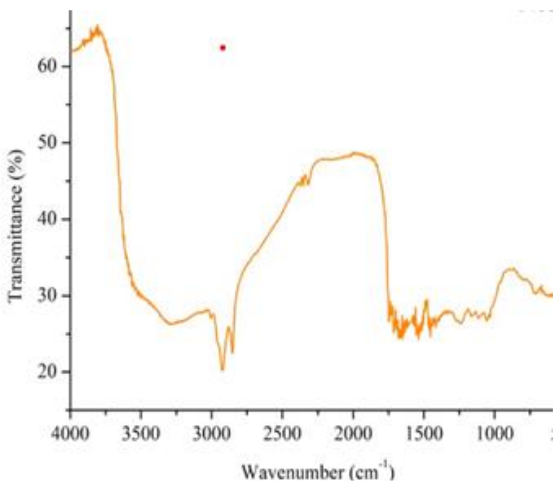

Figure 1. FTIR analysis of papaya seeds

The appearing wavelength of 2850–2970 cm^{-1} indicates the presence of C–H alkane functional group. The peak of the wave that appears at a wavelength of 1690-1760 cm^{-1} indicates the presence of C=O Aldehyde/Ketone/Carboxylic Acid/Ester. The research conducted by Amran,*et al* (2021) in which FTIR was performed on papaya seeds to treat river water showing that papaya seeds show the functional groups of OH, C=O and C–O which are the dominant functional groups in biocoagulants that enable the efficient conversion of proteins into polyelectrolytes. The presence of a large number of functional groups in Papaya seeds indicates its potential to adsorb various contaminants to facilitate the removal of suspended and dissolved substances from water (Amran et al., 2021).

Rahmawati (2016) found that papaya seeds contain polar functional groups such as alcohols, aldehydes, ketones, carboxylic acid, phenol, and ether, these groups are able to bind metal ions by donating an electron pair to form complexes with metal ions in solution. Another characteristics analysis was carried out using Proximate analysis which results can be seen in Table 1.

Table 1. Proximate analysis

Parameter	Concentration (%)
Protein	21.59
Fat	26.32
Water	7.33
Ash	8.13
Carbohydrate	36.63

The results of the proximate analysis showed that papaya seeds contain 21.59% protein, 26.32% fat, 7.33% water, 8.13% ash and 36.63% carbohydrates. The proximate tests conducted by Anggorowati (2021) resulted in the papaya seeds containing 7.3% of water; 30.1% of oil; 28.1% of protein; 8.2 % of ash; and 25.6% of carbohydrates.The water content determines the seeds durability and the requires sample the treatment, while the ash content shows the percentage of the total inorganic or mineral components contained in the sample (Putra & Ainun, 2021). Proteins dissolved in water are positively charged and have properties similar to Alum polyelectrolytes which can bind colloidal particles and turbid water particles and cationic Amino acids to allow precipitate to occur (Yimer and Bayisa, 2021). Proteins and Carbohydrates in natural polymers function as coagulants (Anggorowati, 2021).

3.2. Effect of the Treatment on Turbidity and TSS

The treatment applied in this study consist of doses, particle sizes, and mixing speeds. The initial turbidity and concentration of total suspended solids (TSS) of the wastewater sample are 754 NTI and 124 mg/L), respectively. The turbidity and TSS changes after being treated with papaya biocoagulant are tabulated in Table 2.

Table 2. Effect of treatment on turbidity and TSS

Treatment	Turbidity (NTU)	TSS (mg/l)
D1P1KP1	601.33	20
D1P1KP2	409.33	46
D1P1KP3	438.67	42
D1P2KP1	387.00	49
D1P2KP2	334.00	56
D1P2KP3	355.67	53
D1P3KP1	304.00	60
D1P3KP2	275.33	63
D1P3KP3	308.67	59
D2P1KP1	252.33	67
D2P1KP2	226.67	70
D2P1KP3	242.33	68
D2P2KP1	216.00	71
D2P2KP2	205.33	73
D2P2KP3	209.67	72
D2P3KP1	197.33	74
D2P3KP2	179.33	76
D2P3KP3	188.33	75
D3P1KP1	173.00	77
D3P1KP2	160.67	79
D3P1KP3	170.00	77
D3P2KP1	151.67	80
D3P2KP2	141.00	81
D3P2KP3	145.67	81
D3P3KP1	136.33	82
D3P3KP2	92.40	88
D3P3KP3	131.67	83

Remarks:
D1: Dose of 250 mg
D2: Dose of 500 mg
D3: Dose of 750 mg
P1: 60 mesh
P2: 120 mesh
P3: 230 mesh
KP1: 120 rpm & 10 rpm
KP2: 180 rpm & 30 rpm
KP3: 200 rpm & 50 rpm

The results show that the doses, particle sizes, and mixing speeds simultaneously produced the lowest turbidity in D3P3KP2 (dose of 750 mg, particle size of 230 mesh, and mixing speed of 180 rpm & 30 rpm). The highest turbidity was found in D1P1KP1 (dose of 250 mg, particle size of 60 mesh, and mixing speed of 120 & 10 rpm) with turbidity of 601.33 NTU. The highest TSS was found in D1P1KP3 (dose of 250 mg,particle size of 60 mesh, and mixing speed of 200 rpm & 50 rpm). The lowest TSS was found in the D3P3KP2 (dose of 750 mg, particle size of 230 mesh, and mixing speed of 180 rpm & 30 rpm). Increasing the dose of the coagulant causes more suspended particles to be entangled in hydroxide precipitates which causes higher colloid removal (Asharuddin et al., 2018). The smaller the particle size of the biocoagulant, the more turbidity can be reduced.

Particle size affects the performance of the coagulant because the smaller the size of the coagulant particles, the greater the surface area of the coagulant which can make the aggregation process between coagulants and particles in solution easier to occur, so the potential for the aggregation process between solid particles and coagulants will be greater and cause flocs are easier to form (Putra & Ainun, 2021). The floc from the coagulation-flocculation process is brittle and easily broken. Defective floc will reduce removal efficiency and increase pollutant concentration in wastewater. Meanwhile, overly high mixing speed can damage what is formed and increase the concentration of pollutants in the wastewater (Britza et al., 2006).

3.3. Sludge Volume Index (SVI) and Sludge Mass (SM)

The SVI and SM analyses were performed to evaluate the settling ability of the coagulation process. The SVI is one of the parameters to determine the characteristics of the sludge and its settling performance. The SM is used to determine residuals that forms into precipitates following the addition of the coagulants. The SM shows the ability of coagulants to bind particles so that they precipitate out in the form of residues that can be separated by filtering (Asharuddin et al, 2018). The data of SVI and SM are shown in Table 3.

The results showed that the treatment with the highest SVI of 86.13 ml/g and the lowest SM of 81.48% is D3P3KP2 (dose of 750 mg with a particle size of 230 mesh and a mixing speed of 180 rpm & 30 rpm) and the treatment with the lowest SVI of 32.70 ml/g and the highest SM of 82.95% is D1P1KP1 (dose of 250 mg, particle size of 60 mesh, and mixing speeds of 120 rpm & 10 rpm). Based on these data when the SVI is high, the SM is low. Low SM will give a higher Sludge Volume Index (Clifton, 1998). SVI indicates the ability of colloids in water to precipitate, the greater the SVI, the better the colloid's ability to precipitate (Yimer and Bayisa, 2021). SVI values of below 80 mL/g are considered very good, while the values between 80 – 150 ml/g are moderate, and the values between 76 – 80 ml/g indicate a good sedimentation ability (Diaz et al., 2017).

Table 3. Sludge Volume Index and SM

Treatment	SVI (ml/g)	SM (%)
D1P1KP1	32.70	82.95
D1P1KP2	33.24	82.85
D1P1KP3	36.69	82.84
D1P2KP1	37.95	82.86
D1P2KP2	35.73	82.83
D1P2KP3	37.83	82.83
D1P3KP1	38.41	82.77
D1P3KP2	38.89	82.78
D1P3KP3	38.77	82.77
D2P1KP1	47.32	82.65
D2P1KP2	48.54	82.56
D2P1KP3	48.28	82.40
D2P2KP1	51.21	82.55
D2P2KP2	53.82	82.51
D2P2KP3	50.61	82.48
D2P3KP1	52.70	82.33
D2P3KP2	52.30	82.44
D2P3KP3	52.77	82.42
D3P1KP1	68.46	82.35
D3P1KP2	70.96	82.38
D3P1KP3	68.87	82.40
D3P2KP1	69.10	82.37
D3P2KP2	73.12	82.39
D3P2KP3	72.18	82.36
D3P3KP1	73.11	82.33
D3P3KP2	86.13	81.48
D3P3KP3	72.69	82.32

Remarks:
D1: Dose of 250 mg
D2: Dose of 500 mg
D3: Dose of 750 mg
P1: 60 mesh

P2: 120 mesh
P3: 230 mesh
KP1: 120 rpm & 10 rpm
KP2: 180 rpm & 30 rpm
KP3: 200 rpm & 50 rpm

The SM is one of the parameters to determine the ability of natural coagulants because it shows the mass of precipitate formed in from the coagulation process. The lower the produced precipitate, the better the performance of the coagulant (Diaz et al., 2017). Increasing the coagulant dose result in, greater aggregation between particles and better particle destabilization (Putra & Airun, 2021). Particle size influences the floc during the flocculation coagulation process. The smaller the particle size, the larger the surface area, and the wider the absorption surface (Novita et al., 2019). The flocs formed from this process affect the SVI and SM values (Airun, 2020). The stirring process helps mix the coagulant with the water, destabilize the particles, and combine the precipitates that form into floc, hence the mixing speed causes the formation of larger flocs (Lin et al., 2013).

4. Conclusion

Papaya seed biocoagulant has functional groups potential for absorbing various contaminants to facilitate the removal of suspended and dissolved substances from the wastewater. D3P3KP2 (dose of 750 mg, particle size of 230 mesh, and mixing speed of 180 rpm & 30 rpm) can reduce turbidity from 754 NTU to 92 NTU and TSS from 124 mg/L to 79 mg/ L. D3P3KP2 has the highest SVI (86.13 ml/g) and the lowest SM (81.48 %). Therefore, it can be concluded that to reduce the use of chemical coagulant for treating wastewater from dairy industry papaya seeds can be used as biocoagulant.

Author Contributions

Conceptualisation, Z.R.; Formal analysis, Z.R.; Investigation, R.W., Z.R.; Project administration, Z.R.; Supervision, B.S.; Writing – original draft, Z.R.; Writing – review and editing, B.S.

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Data Available Statement

Available on request to the corresponding author.

Conflict of Interest

The author declares that there is no conflict of interest regarding the publication of this manuscript.

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