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Effectiveness of Green Mussel Shells as a Natural Coagulant for Domestic Wastewater Treatment by Jar Test

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ABSTRACT Coagulation is one of the wastewater treatment processes. Aluminum sulfate is usually used as a chemical coagulant. On the other way, the use of alum can cause a new problem that was the inorganic residues produced are carcinogenic and can harm the environment and human health. Then people try to use natural materials such as chitosan as the coagulant. Chitosan can be produced from the skin of marine animals such as green mussels. This study aimed to determine the effectiveness of natural coagulant green mussel shells. The research method was an experimental study by testing the effectiveness of green mussel shells as a natural coagulant with one group post design. This research used small dose of chitosan that is 0,15 g/l until 0,4 g/l to know which one is the most effective to reduce turbidity and suspended solid. The sample in this study was domestic wastewater. The parameters were total suspended solids (TSS) and turbidity. The data were analyzed by the ANOVA test. The result showed that both TSS and turbidity were decreased by about 100% (14 mg/l to 0,65 mg/l) for optimum decreasing of TSS parameter and 73% (2.38 NTU to 0 NTU) for turbidity parameters. The optimal dose of natural coagulant green mussel shells for domestic wastewater was 0,4 g/l. The ANOVA test showed that there was an effect of green mussel shells coagulant dose on TSS and turbidity level (p-value = 0,000). The potential of using these green mussel shells as a coagulant is indeed very high. However, it takes a combination with other natural ingredients to make an effective and safe coagulant.

INDEX TERMS coagulation, green mussel shells, chitosan.

I. INTRODUCTION

Water is a very important thing for life because it is used in various daily activities such as drinking, bathing, and washing needs. Water is also used in other activities such as agriculture, livestock and industry. But now, water contamination becomes a big problem for human that water can't be used properly. Water contamination can be caused by domestic waste stairs, industry, agriculture, and others. Domestic wastewater becomes the biggest source of water contamination in Indonesia [1]. Domestic wastewater is the water that has been used by a community. Domestic wastewater contains materials from human activities such as personal washing and laundry[2].

Domestic wastewater can be divided into two, namely grey water and black water. Grey water is domestic liquid waste originating from washing activities such as washing dishes, water used for washing dishes, bathing, and washing clothes [3]. While black water is liquid waste from toilets and septic tanks. Untreated grey wastewater directly dumped into the water body. It is danger to the environment. So that it can pollute the river and be the place where the ditch empties. As a result, the river turns brown as well emit an unpleasant odor. In addition, it can cause fish those in the river died. Ingredient pollutants contained in the waste can also be a source of disease, such as cholera, dysentery, and various other diseases [4].

Generally, people throw the wastewater to the environment without process it first. They assume that domestic waste thrown directly into the sewer will not have a negative impact. Pollution of the aquatic environment increases along with the increase in population and industrial development. The presence of hazardous chemical compounds in the waters contributes to pollution and harms health. Treatment of wastewater has become important to protect the environment and human health[5].

There are many technologies to treat domestic wastewater. Aerated biological methods is normally used to treat domestic wastewater because it can produce high quality effluent. Activated sludge, one of biological treatment, can remove almost 90% biological oxygen demand (BOD) and suspended solids (SS) [6]. This treatment also has disadvantage, that is requiring continuous air supply, high operating cost, and sludge disposal. Other treatments can also be used to remove pollutant in wastewater, such as filtration, sedimentation, coagulation, and flocculation. These treatments are commonly used physical and chemical processes to remove suspended material in wastewater [7].The domestic wastewater treatment process consists of several processing stages. One of them is the coagulation process. Coagulation-flocculation is one method of handling liquid waste using coagulant or flocculant material to produce colloid destabilization and the formation of flocs to form agglomerations. Coagulant materials can use organic or inorganic compounds. Common coagulants commonly used are ferric chloride, ferric sulfate, and aluminum sulfate ($Al_2(SO_4)$)[8].

The coagulation process mostly uses chemicals such as aluminum sulfate (alum). Alum is a chemical that is often used as a coagulant. Even alum can remove microplastics in water [9]. The use of alum as a coagulant raises concerns because the inorganic residues produced are carcinogenic [10]. In addition, the use of alum can cause effects on the environment and health because this type of coagulant is not easily biodegradable. This is what encourages researchers to look for alternative coagulants, especially those from natural ingredients.

Continuous use of synthetic coagulants will certainly have a negative impact because they will accumulate in the body. Several studies have proven that wastewater can be purified using natural coagulants such as moringa seeds, tamarind seeds, and winged beans [11]. The use of natural materials needs to be developed so that it can reduce residues from chemicals that have been used so far. One of the natural ingredients that can be used as a coagulant is chitosan. Chitosan can be obtained from various materials. The chitosan used in this study is from green mussel shells.

Chitosan is a type of natural polymer with chain straight and has the general formula ($C_6H_{11}NO_4$) otherwise known as poly(β-(1-4)-2-amino-2-deoxy-Dglucopyranose) is a bio-polymer that contains free amino groups and the hydroxyl group on the carbon chain so that it makes chitosan reactive. Chitosan has properties biocompatibility, biodegradation, no toxic, non-allergenic and capable of fiber formation and films. In addition, chitosan also has absorbing properties good agglomerate, this property can increase reactivity in derivative manufacture [12]. Chitosan comes from mussel shells waste which contains chemical compounds in the form of chitin, which is used as an absorbent to absorb heavy metals such as zinc (Zn), chromium (Cr), copper (Cu), cobalt (Co), nickel (Ni and Iron (Fe) on a lab scale Given the enormous benefits of chitosan compounds and the availability of raw materials that are plentiful and easy to obtain. It is necessary to study and develop this waste as an absorbent material for heavy metals in water. It has the ability as a coagulant because it contains a lot of nitrogen in the amine group. The amine and hydroxyl groups make chitosan more active and polycationic, these properties are used as a coagulant in peat water treatment which can absorb more Fe metal than Poly-aluminum chloride[13]. Chitosan can be made from any crustacea, such as shrimp and crab. These chitosan remove 100% of color from textile industry effluent [14]. Crustacean's shelss contained chitosan is potential to be nature coagulant to remove total suspended solid, BOD, and COD [15].

The purpose of this study was to test the effectiveness of green mussel shells as a natural coagulant in domestic wastewater treatment. The coagulant obtained from green mussel shell chitosan is expected to be an environmentally friendly material and has a high added value. This study used green mussel shells from Surabaya beach which was contained may organics matter. Public can imply this chitosan to clean their domestic wastewater.

II. METHODOLOGY

A. DESIGN OF STUDY

Coagulation-flocculation is one method of handling liquid waste using coagulant or flocculant material to produce colloid destabilization and the formation of flocs to form agglomerations. One of coagulant that can be used is chitosan. The design of this study was experimental - one group post design. The location of this research was conducted at the Laboratory of the Department of Environmental Health Poltekkes Kemenkes Surabaya. The object of this research is domestic wastewater originating from household activities. The independent variable was the dose of green mussel shells given to domestic wastewater. The doses used were 0.15 g/l, 0.20 g/l, 0.25 g/l, 0.30 g/l, 0.35 g/l, and 0.40 g/l. The dependent variables were the turbidity and TSS parameters obtained from the coagulation process of green mussel shells.

B. THE PROCESS OF MAKING CHITOSAN

The process of making chitosan is as follows and as presented in figure 1. Wash the green mussel shells first, then reduce the size of the green clam shell. Sieve the green mussel shells with a 80 mesh. After that, the mussel shells need to be deproteinization, demineralization, and deacetylation processes with strong acid and base solutions. Dry the results of the above process and the product is ready for the jar test.

FIGURE 1 Chitosan Making Processes

The wastewater sample used in this study is domestic waste originating from household activities in the community in Kertajaya Village, Gubeng District, Surabaya City. The green mussel shells used were obtained from the Kenjeran area, Surabaya City, East Java Province as presented in figure 2. The coagulant size used is 80 mesh. The domestic wastewater sampling was according to SNI. 6989.59.2008 with the time composite method. Point sampling is carried out at the flow outlet household wastewater before entering to the drainage.

FIGURE 2 **Green Mussels Shells**

FIGURE 3 **Green Mussels Shells Chitosan**

The manufacture of chitosan from green mussel shells was divided into 3 stages. The first stage is the deproteination stage using 3% NaOH 1:6 (w:v) and heated at 85 °C for 30 minutes. Furthermore, this mixture was cooled and filtered, the filtered residue was washed with distilled water until neutral and then dried in an oven at 20 °C for 24 hours. The second stage is demineralization using 1,25 N 1:10 (w:v) HCl solution and heated at 75 °C for 1 hour. The reaction product was filtered and washed with distilled water until neutral. Then it was dried in an oven at 20 °C for 24 hours. The last step is deacetylation, the isolated chitin is then removed from the acetyl group with 45% 1:20 (w:v) NaOH solution and heated at 140 °C for 1 hour. The result is filtered and washed with distilled water until neutral. Finally, the chitosan was dried in an oven at 80 °C for 24 hours. After this stage, the green mussel shell powder which has become white chitosan powder is ready to be used as a natural coagulant to treat domestic wastewater as presented in figure 3 [16].

C. *JAR TEST*

Domestic wastewater samples were taken from the community. Green mussel shells powder are put in a glass beaker with the following doses 0.15 g/l, 0.20 g/l, 0.25 g/l, 0.30 g/l, 0.35 g/l, and 0.40 g/l. Then the mixing process was carried out by doing fast stirring at 100 rpm for 60 seconds and slow stirring at 60 rpm for 30 minutes, then allowed to stand for 60 minutes to form a precipitate. The precipitated solution was then examined in the laboratory to measure the levels of TSS and turbidity. This method is called jar test as presented as figure 4.

Observations were made during the coagulation process, starting from the process of mixing coagulant in the domestic wastewater sample to the sedimentation process of the formed floc. The process of mixing the coagulant in the sample water was carried out at a speed of 100 rpm for 60 seconds, then at a low speed of 60 rpm for 30 minutes, and allowed to stand for 60 minutes to form a precipitate. Examination of TSS levels and turbidity in sample water before and after coagulation with coagulant doses of green mussel shell powder 0.15 g/l, 0.20 g/l, 0.25 g/l, 0.30 g/l, 0.35 g/l, and 0.40 g/l were carried out at the Surabaya Health Laboratory Center. Examination Turbidi levels using Portable Turbidimeter TURB 355 IR and TSS levels using gravimetri methode.

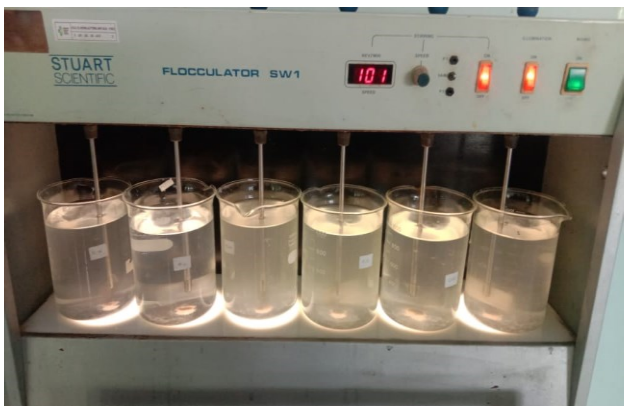


FIGURE 4 **Jar Test**

D. *EFFECTIVITY TEST*

The test aims to determine the difference in the reduction of TSS levels and turbidity in domestic wastewater. The requirements used for the ANOVA test are that the data are normally distributed using the normality test. If the data is normally distributed, then proceed with the homogeneity test to know the difference between the two groups with different data sources. If the data is homogeneous then proceed to the ANOVA test to know whether there is an average difference in the two data groups with a significant value <0, 05. The effectiveness test for reducing TSS and turbidity levels is calculated using the following Eq. (1):

$$\text{Effectivity (Ef)} = \frac{(co-ct)}{co}\ x\ 100\% \ \ldots\ldots\ldots\ldots\ldots\ldots\ldots (1)$$

Note:
Ef : The effectiveness of green mussel shell powder as a natural coagulant
Co : TSS concentration and turbidity before coagulation
Ct : TSS concentration and turbidity after coagulation

III. RESULT

The chitosan has some characterization as presented in TABLE 1:

TABLE 1
The Characterization of Chitosan

No	Parameters	Unit	Standard SNI 7949:2013	Result
1	Colours	-	Light brown to white	Brownish White
2	Water content	%	≤12	5
3	Metal content	%	≤1	0,085
4	Degree of deacetylation	%	≥ 75	83,05

Measurement of TSS and turbidity levels before the coagulation process using green mussel shell powder was carried out to determine the initial characteristics of the domestic wastewater used as presented in table 2. Furthermore, a comparison is made with the Minister of Environment Regulation No. 68 of 2016. The results of the TSS and turbidity measurements are as shown in TABLE 2:

TABLE 2
TSS and Turbidity Level before Coagulation using Green Mussel Shell

No	Parameters	Unit	Standard	Result
1	TSS	mg/l	30	14
2	Turbidity	NTU	5	2,38

Table 2 shows that TSS and turbidity level in domestic wastewater before the coagulation process were 14 mg/l and 2.38 NTU. This result showed that TSS and turbidity level was still under the standard. The domestic wastewater was being treated by green mussel shells coagulant. The results of the TSS and turbidity measurements are as shown in TABLE 3:

TABLE 3
TSS and Turbidity Level after Coagulation using Green Mussel Shell

No	Parameters	Unit	Standard	Result
1	TSS	mg/l	30	0
2	Turbidity	NTU	5	0,65

Based on TABLE 3 TSS and turbidity levels in domestic wastewater after the coagulation process are 0 mg/l and 0.65 NTU. This result showed that there were differences between before and after coagulation using green mussel shells. Analysis of the Effectiveness of Green Mussel Shells as a Natural Coagulant in Reducing TSS and Turbidity Levels in Domestic Wastewater are as follows TABLE 4:

TABLE 4
Analysis of the Effectiveness of Green Mussel Shell Coagulant on TSS Levels in Domestic Wastewater

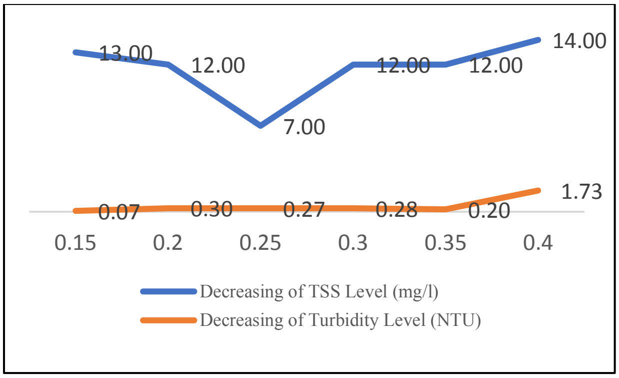
No	Coagulant dose (g/l)	TSS Level (mg/l)		Effectiveness (%)
		Before treatment	After treatment	
1	0,15	14	1	93%
2	0,20	14	2	86%
3	0,25	14	7	50%
4	0,30	14	2	86%
5	0,35	14	2	86%
6	0,40	14	0	100%

Based on TABLE 4, it can be seen that in general there was a decrease in TSS levels after being added with green mussel shell powder coagulant between 1 - 7 mg/l. The lowest percentage decrease in TSS levels was at a dose of 0.25 g/l by 50% and the highest decrease in TSS levels at a dose of 0.40 g/l was 100%.

TABLE 5
Analysis of the Effectiveness of Green Mussel Shell Coagulant on Turbidity Levels in Domestic Wastewater

No	Coagulant dose (g/l)	Turbidity Level		Effectivenes s (%)
		Before treatmen t	After treatmen t	
1	0,15	2,38	2,31	3%
2	0,20	2,38	2,08	13%
3	0,25	2,38	2,11	11%
4	0,30	2,38	2,1	12%
5	0,35	2,38	2,18	8%
6	0,40	2,38	0,65	73%

Based on TABLE 5, it can be seen that the turbidity level generally decreased after being added with green mussel shell powder coagulant between 0.07 – 1.73 NTU from the initial level of 2.38 NTU. The lowest percentage decrease in turbidity levels at a dose of 0.15 g/l was 3% and the highest decrease in turbidity levels at a dose of 0.40 g/l was 73% as presented in figure 5.



TSS level (mg/l) and Turbidity Level (NTU)

FIGURE 5 Decreasing of TSS and Turbidity Level After Coagulation

FIGURE 5 shows that the highest decrease in TSS and turbidity levels was at a dose of 0.4 g/l.

TABLE 5
ANOVA Test Result

No	Parameters	Sig p-value
1	TSS	0,049
2	Turbidity	0,000

Based on the results of the ANOVA test (TABLE 5), there was an effect of the dose of green mussel shell powder as a natural coagulant on TSS and turbidity levels in domestic wastewater with a significance value of 2-tailed TSS of 0.049 and turbidity of 0.000.

IV. DISCUSSION

This study used chitosan from green mussel shells. The chitosan was made from green mussel shells at Surabaya beach which is rich of organic matter. This study was only used domestic wastewater as the object since the domestic wastewater become a serious problem [17]. Chitosan is a polymer obtained by changing the group acetamide (-$NHCOCH_3$) in chitin to the amine group (-NH_2). Therefore, the release of the acetyl group on chitin acetamide produces a deacetylated amine group. Determination of the degree of deacetylation using the baseline method on the results of the FTIR analysis [18]. Ability to form floc by coagulant chitosan depends on the degree of deacetylation. The higher the degree of deacetylation, the lower the acetyl group in chitosan, so that the interactions between ions and hydrogen is getting stronger[19].

Jar test is used to test the effectivity of the coagulant. Jar test carried out with rapid stirring and slow stirring. It is intended that floc can be formed optimally. Rapid stirring aims to disperse coagulant until homogeneous with a short time, while slow stirring aims to prevent so that the floc core does not break and contact between the floc cores so that it is easier to deposited[20]. A jar test was conducted as a batch process and consisted of a multiple flocculator system. The domestic wastewater sample used for this study was taken from the liquid waste of the residential area of Kertajaya, Gubeng, Surabaya. Those raw water was poured into 1-L beakers of 4 unit. Rapid mixing of 150 rpm for 1 minute and slow mixing of 35 rpm for 15 minutes. The floc formed was allowed to settle for 30 minutes after terminating the agitation [21].

The results of the laboratory examination showed that the TSS level was 14 mg/l and the turbidity was 2.38 NTU. The results stated that the TSS level in the domestic wastewater had exceeded the domestic wastewater quality standard stipulated in the Regulation of the Minister of Environment and Forestry of the Republic of Indonesia No. 68 in 2016 which is 30 mg/l for TSS.

Turbidity in water can be caused by the presence of suspended solids in water, both organic and inorganic substances. Substance inorganic, usually in the form of weathered rock, sand, silt, and dissolved metals. Whereas organic matter comes from domestic waste and industrial that can used as food for bacteria and bacterial proliferation. Microorganisms, algae, and plankton can also causes turbidity of the water [22].

TSS or total suspended solids are suspended materials or particles having a diameter of >1 m that are retained on a milli pore with a pore diameter of 0.45 m. Domestic wastewater tends to have a fairly high TSS value because it comes from various activities such as bathing, washing, and other activities that use water. The presence of TSS in water can increase the value of turbidity and can inhibit the penetration of sunlight into the water column and ultimately affect the photosynthesis process in the waters[23]. Domestic waste that contains TSS and turbidity is quite high and it is directly dumped into a ditch without being treated. domestic wastewater is collected in underground pipes which are called “sewers”. The wastewater in sewers flows by gravity. Untreated wastewater causes major hazard to the environment and to the human health [2].

The results showed that TSS and turbidity levels after adding natural coagulant from green mussel shell powder decreased most optimally at a dose of 0.4 g/l of 14 mg/l for TSS levels and 1.73 NTU for turbidity levels. The maximum decrease in TSS levels and turbidity was at a dose of 0.4 g/l. TSS and turbidity levels are below the wastewater quality standard. The decrease in TSS and turbidity levels was due to the rapid stirring of bio-coagulants in the sample water, thus helping in the process of mixing the coagulant material evenly in the sample water (domestic waste). The coagulant that has been dispersed in the wastewater will be able to bind more suspended solids and produce more sediment.

The decrease in TSS levels in the treatment of 0.4 g/l chitosan was due to the attachment of chitosan molecules to the colloidal surface which would cause the chitosan to have relatively high chemical reactivity is quite high and this can cause the poly-electronic nature of the cation so that it acts as an absorbent on wastewater. In other words, chitosan is non-toxic and capable of producing flocs that will settle with the particles in the wastewater, so this will reduce the negative effects to aquatic life. This result showed that green mussel shell powder is effectively used as a bio-coagulant. The optimum dose to reduce the levels of TSS and turbidity is 0.4 g/l with the percentage reduction for TSS being 100% and turbidity being 73%. Some factors that affect the effectivity of coagulation is coagulant dose, detention time, and speed of slow and rapid stirring. Based of research of chitosan for harvesting Microalgae Spirulina sp, the optimum condition for chitosan as a flocculant was achieved at a slow stirring speed of 40 rpm, sedimentation time of 2 hours, chitosan concentration of 100 mg/l and pH of 7-8 [24]. Chitosan from green scallop shells are also effective to remove COD, BOD, TSS levels in tofu industrial wastewater [25]. Coagulation by chitosan, alum, and PAC were all optimum within a range oh neutral pH [26].

Small dose of coagulant can inhibit coagulation process that is being lacking in floc cores, leaving more colloidal particles, so that the higher the dose of coagulant given, the colloidal particles combined to form more macrophages and leave fewer colloids in the final product [27]. However, administration of a coagulant dose that is too large can also cause the water to become cloudy because it can inhibit the floc formation process due to too many cations which causes the electrostatic force on the colloid that has been fused to the macro flocs to become large and results in the destruction of the bonds that have been formed. Fast stirring during the jar test process helps the mixing process coagulant in wastewater equally. The coagulants have been dispersed in the wastewater will bind suspended solids more, therefore it will the results obtained from the precipitate of solids better suspension. But at larger doses there will be a decrease in speed continuous deposition slow. The addition of a dose that is too large resulting in an excess of positive ions produce a large enough repulsion which causes movement particles in the water and interfere with the process the stabilization that has occurred. This can cause binding failure and floc formation [28]. The flocculation coagulation process carried out to eliminate a number of particles initially present in wastewater. As a result, the turbidity measurement blocking light entering water becomes less and less so the measured turbidity becomes increasingly small. In other words, reduced particles after the process of causing the turbidity of the water is also getting reduce.

TSS levels can fluctuate due to the stabilization of colloidal particles due to the added dose. Re-stabilization is the process of reversing the charge of colloidal particles which are generally negatively charged and turn positive due to the absorption of an excess dose which results in a repulsive force between colloidal particles because they have the same charge[29]. This causes a larger floc cannot to be formed and the TSS content increases in the sample water [30]. The turbidity of the sample after going through the coagulation process has a trend that is almost the same as the decrease in TSS. However, the process of decreasing TSS and turbidity are not always linearly related. Smaller TSS levels will not necessarily have a small turbidity value because several other factors can cause turbidity besides suspended solids. Limitation of weaknesses of this research are the source of the chitosan was from one location and the indicator to know the effectiveness of this chitosan was only two indicators. It needs advance research to add the source of the green mussel shells and the indicators of the effectiveness.

V. CONCLUSION

Green Mussel Shells has been proven to be effective in reducing TSS and turbidity level in domestic wastewater. The results showed that TSS and turbidity levels after adding natural coagulant from green mussel shell powder decreased most optimally at a dose of 0.4 g/l of 14 mg/l for TSS levels and 1.73 NTU for turbidity levels. It is necessary to carry out the next stage of processing (filtration) to produce better quality wastewater by following domestic wastewater quality standards. Further research is needed on the combination of other natural ingredients as coagulants for domestic wastewater treatment process.

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