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A Tofu Wastewater Treatment Using Combination of Plasma Electrolysis and Coagulation-Flocculation Method

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Abstract: In this research, tofu wastewater was treated using a combination of plasma electrolysis and coagulation-flocculation with a 1% chitosan solution as the coagulant. Untreated tofu wastewater is an organic waste with very high COD and TSS, 7771.3 mg/L and 840.5 mg/L, respectively. Coagulation-flocculation method was used for wastewater pretreatment because it could reduce the high pollution parameters in tofu wastewater while plasma electrolysis could reduce them to their smallest concentration. The best chitosan volume for reducing TSS to 74.4% was 20 mL, while the best volume for lowered COD to 16.8% was 40 mL. Plasma electrolysis was the next step for tofu wastewater degradation; it could drop the value of COD of tofu wastewater to 36% during a 60-minute process at 2000 mg/L as the initial concentration of the wastewater. This methodology reduced the environmental damage caused by the wastewater with an effective treatment.

Keywords: *Coagulation-flocculation, plasma electrolysis, TSS, COD*

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I. INTRODUCTION

In 2014, 954.557 tons of soybeans were produced in Indonesia, and according to data from the Ministry of Agriculture, the amount of soybeans imported in the same year were almost 2 million tons. [**?**] mentioned that 38% of soybean in Indonesia is consumed in the form of tofu, but unfortunately, as much as 1 kg of soybean can produce pollution load of BOD as much as 80 ± 5 gram, dissolved COD 80 ± 20 gram, and TSS 9 ± 3 gram [**?**].

Tofu wastewater has some high pollution parameters; the value of those can be seen in Table **??**.

TABLE 1
POLLUTION PARAMETERS IN TOFU WASTEWATER

Parameter	Untreated tofu wastewater
pH	3.9
COD (mg/L)	7771.3
TSS (mg/L)	840.5

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Untreated tofu wastewater requires pretreatment before further processing [?]. Pretreatment is done so that the significance of the pollution parameters degradation presented in the waste can be better [?]. In this study, coagulation-flocculation using coagulant chitosan as a pretreatment was conducted. The flock formed by the addition of coagulant would settle, so that the total amount of Suspended Solid (TSS) and Chemical Oxygen Demand (COD) in the waste could be decreased [?].

Besides, the precipitates obtained from coagulation using chitosan can be used as feed additives since they contain no inorganic materials, such as iron and aluminum. Chitosan as an additional ingredient in the feed, has been approved by the US. Food and Drug Administration Knorr, 1984 in [?].

The main wastewater treatment in this study is plasma electrolysis. Plasma electrolysis is a breakdown of conventional analysis. Many factors have affected plasma electrolysis, such as: electrode material, electrolyte solution, and the nature of reaction in anode [?].

Many researches have proved that coagulation-flocculation was good to be used for reducing high concentration of pollution parameters in organic wastewater [? ?] while plasma electrolysis, with the reactive species, could be used to degrade the pollutants until their small concentrations [? ?]. So, we combined those two methods to degrade high pollution parameters in tofu wastewater effectively, such as, TSS and COD.

A. *Objective of the Study*

To obtain the concentration of chitosan solution required in the coagulation-flocculation process and to achieve the best initial concentration of tofu wastewater to be degraded in plasma electrolysis.

II. LITERATURE REVIEW

A. *Tofu Wastewater*

Tofu wastewater has several physical properties, organic contents and inorganic contents. Some physical properties contained in the liquid waste are:

1) Total solids: The total solids content (total solid) in the liquid waste is the sum of all the material remaining as residue after evaporation at a temperature of 103-105°C. Materials lost during evaporation are not categorized as solid. The residue obtained after evaporation can be classified as suspended solid or filterable solid. Suspended solid is a solid that does not pass the filtering process through a filter. The filter used is usually a glass-fiber filter with a pore size of 1.2 micrometers [?]. The value of TSS in tofu liquid waste is 500 $\pm$ 250 mg/L [?].

2) Temperature: Tofu wastewater temperatures range from 37°C to 45°C [?]. The temperature of this tofu liquid waste comes from the tofu molding process at 30-35°C and the soybean cooking process at 80-100°C [?].

3) Color: The colored wastewater can absorb more oxygen in the water, which will make the water become black and smelly for a long time. The color of tofu wastewater amounted to 2.225-2.250 Pt.Co [?].

4) Turbidity: The level of turbidity of the tofu wastewater can be measured by the ratio between the intensity of light dissipated by the waste-water and the intensity of light dissipated by the standard suspension under the same conditions. The turbidity level has no relationship with the suspended solid concentration in untreated liquid waste [?]. According to [?], the level of turbidity of tofu wastewater ranges between 535 and 585 FTU.

5) Odor: Many tofu industries are unacceptable to the public due to the smells that arise. The foul smell produced in tofu industry waste is the result of organic material decomposition in tofu wastewater [?].

Tofu wastewater also has some chemical components. They are:

6) Organic materials: Organic ingredients contained in tofu are proteins, carbohydrates, and fats. Proteins are present in complex chemical structures and decompose easily into many forms. Protein is one source of nitrogen in liquid waste. The largest organic ingredient in tofu liquid waste is protein, as much as 40-60% in the wastewater [?].

Carbohydrates in wastewater can be present as sugar, starch, cellulose, and wood fiber. Carbohydrates contain carbon, hydrogen, and oxygen. Some carbohydrates, like sugar, are water-soluble, while other carbohydrates, such as starch, are insoluble in water. Carbohydrate content in tofu liquid waste ranges from 25 to 50% [?].

Another organic material in tofu wastewater is fat. Fat is a stable organic component which is not decomposed easily by bacteria. Fat is an ester compound of alcohol or glycerol (glycerin) with fatty acids. The fat content in tofu waste is only 10% [?].

7) Inorganic materials: Inorganic compounds present in the tofu wastewater are generally acidic. The total nitrogen compounds are comprised of organic nitrogen, ammonia, nitrite, and nirate. Organic nitrogen can be determined using the Kjeldahl method. Ammonia is being in the waste as ammonium or ammonia ions, depending on the pH of the waste itself. If the pH of waste is below 7, then the ammonium ion dominates but if the pH of wastes is above 7, then the ammonia is dominating.

The amount of nitrite and nitrate can be determined by using colorimetric method. Nitrite is generally unstable and easily oxidized to nitrate [?]. According to [?], levels of ammonia in tofu wastewater are equal to 23.3-23.5 mg/L.

8) Gas: The gases contained in the tofu waste are the burning fumes and the foul odor resulting from the decomposition of the tofu wastewater. The gases commonly found in tofu wastewater are Nitrogen gas (N_2), Oxygen (O_2), Carbon Dioxide (CO_2), Hydrogen Sulfide (H_2S), Ammonia (NH_3), and Methane (CH_4) [?].

B. Coagulation-Flocculation

Colloids are stable in waste because it has electric charges. Colloidal charges can be positive or negative. However, generally colloidal particles in waste are usually negatively charged [?]. To remove colloids, these particles must be destabilized so that larger and heavier flocs may be formed. These flocs can be removed by coagulation, flocculation, and precipitation [?]. Chitosan, as a polymer, can destabilize this colloid through bridging formation. Too many polymers which are incorporated in the coagulation-flocculation process could make the particles re-dispersed, with a greater amount of positive charge than the negative charge [?]. Several studies have been conducted to see the effectiveness of chitosan in waste treatment. A study conducted by [?] showed that a chitosan ratio with TSS in a biscuit wastewater of 0.45 can decrease TSS in wastewater by 94.44%. TSS and turbidity of tempe industry waste could also be reduced by 95.10%-95.82% and 94.33%-95.17% respectively by using chitosan [?], while turbidity in tofu wastewater can be degraded to 97% by using the same coagulant [?].

C. Plasma Electrolysis

Plasma electrolysis is an electrical process in which plasma can form between one electrode and an electrolyte surface. This method can be used to oxidize various organic pollutants. Organic compounds, such as phenols, could be oxidized using this method successfully [?].

Feed concentration is one of the factors that influence the degradation of organic compounds in plasma electrolysis process. The amount of hydroxyl radical required to oxidize the feed is equal to the amount of hydroxyl radical required for H_2O_2 formation. According to [?], the initial rate of H_2O_2 formation will decrease as the initial feed concentration increases. It happens because the decomposition of the feed is initiated by an attack by a hydroxyl radical. As the feed concentration increases, the hydroxyl radical will be consumed more for feed oxidation and fewer are immersed into H_2O_2.

The electrolyte used in the plasma electrolysis process is Na_2SO_4. The higher electrolyte concentrations used result in greater conductivity. This higher conductivity can increase the size of the plasma which is formed and trigger the production of more hydroxyl radicals. The optimum concentration of Na_2SO4 in plasma electrolysis method was 0.02 M [? ? ?].

III. EXPERIMENT

The experimental apparatus used in coagulation-flocculation was laboratory jar test equipment, as shown in Figure **??**, while the setup of plasma electrolysis process comprised step-up transformer, Diode Bridge, slide regulator, manual circuit breaker, multi-meter, and the reactor of plasma electrolysis. The schematic diagram of the experimental setup of plasma electrolysis can be seen in Figure **??**.

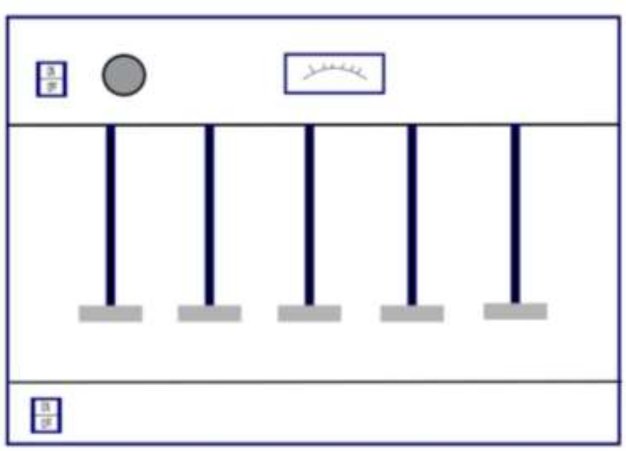

Fig. 1. The schematic diagram of laboratory jar test

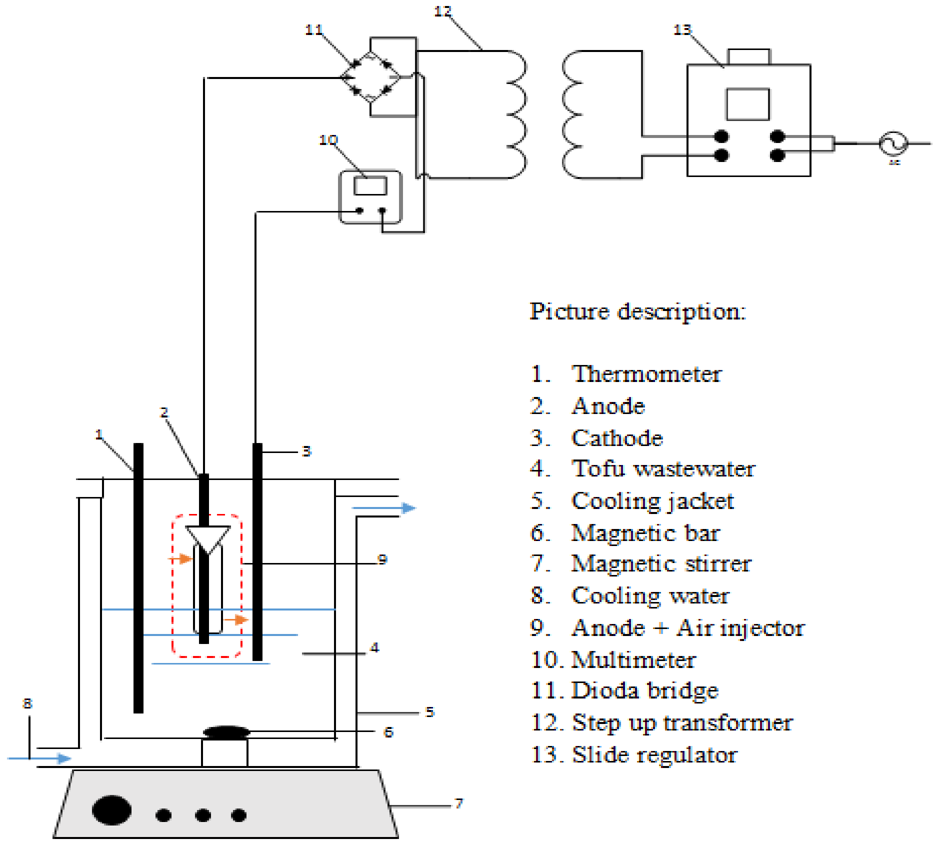

Fig. 2. The schematic diagram of experimental setup for plasma electrolysis

IV. DATA ANALYSIS & DISCUSSION

A. *Coagulation-Flocculation as a Pretreatment of Tofu Wastewater*

The chitosan used in this research was made from small crabs in Indonesia. The degree of deacetylation of the chitosan was about 90%. As much as 5 g chitosan was added to 500 mL of 1% acetic acid solution. The solution was stirred for 2 hours until homogenous.

After the chitosan solutions had been prepared, the laboratory jar test was conducted at 80 rpm as the rapid stirring for 60 seconds. Then, the stirring speed was lowered and allowed to remain constant for 15 minutes at a speed of 30 rpm. The five stirrers, as shown in Figure **??**, were removed from the apparatus, and the solutions were allowed to settle for 60 minutes. This method was taken from AWWA (1977) in [**?**].

Fig. 3. The process of laboratory jar test

There are many aspects that we have to consider in laboratoy jar test, such as: temperature, speed of stirring, pH of the solution, and dosage of coagulant. In this research, we did not vary the temperature and speed of the stirring, because according to [**?** **?**], temperature and speed of stirring had less effect on reducing the pollution parameters in wastewater.

Two parameters which impacted a lot in degrading the pollution parameters in wastewater were pH and dosage of coagulant [**?**]. From the previous study, we know that the optimum pH for treating tofu wastewater was 5.8 [**?**]. This optimum pH was also used in this research. To adjust the pH, we used 0.01 NH_4OH.

As was mentioned above, coagulation-flocculation process was used as a pretreatment to reduce the COD and TSS of tofu wastewater further. From Figure **??**, we know that the best volume chitosan solutions to lower the COD and TSS were, 40 mL and 20 mL, respectively. After those volumes, COD and TSS of the tofu wastewater increased. These enhancement made the colloids in the wastewater restabilized. The excess chitosan would cover the colloids without connecting it with the other colloids, this made colloids fouling the wastewater again [**?**].

The degradation of COD was not as successful as TSS degradation. COD was only reduced by 16.8% while the TSS could be lowered by 74.4%. However, these final results of COD and TSS achieved in coagulation-flocculation process were still high.

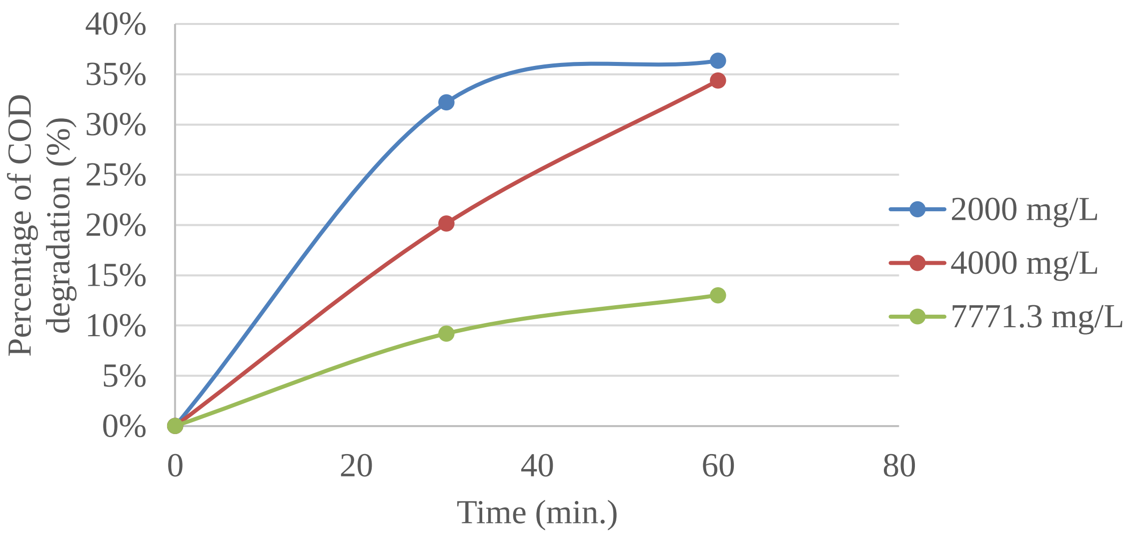

Fig. 4. The effect of volume of chitosan solution on the percentage of COD and TSS degradation in tofu wastewater

B. Plasma Electrolysis in Tofu Wastewater

The next step to reduce COD in this research was by doing a plasma electrolysis. Figure **??** (a) shows the experimental setup for this process. The plasma that was occurred in Figure **??** (b) was kept at high voltage, 700 V, during 30 minutes and 60 minutes with different initial concentrations of the wastewater. The anode and cathode used in this process were tungsten and stainless steel 316, respectively. In addition, we added 40 mg/L $FeSO_4.7H_2O$ as a catalyst and the anode was dipped into the solution as deep as 20 mm. Air was injected into the solution during the plasma electrolysis process. This air injection was used to increase the decomposition rate of wastewater because it produced more OH and HO_2 [**?**]. These radicals could help to reduce the pollution parameters more significantly. Furthermore, according to [**?**], energy consumption was lowered by adding air into the solution.

The electrolyte used in all variations in this process was 0.02 M Na_2SO_4. According to [**?**], 0.02 M Na_2SO_4 was the best concentration of electrolyte to produce the great amount of OH with 20 mm depth of anode. The distance between the anode and the cathode was about 40 mm. [**?**] said that the anode would be destroyed if it was less than 10 mm.

Fig. 5. (a) The experimental setup of the plasma electrolysis (b) Plasma that occurred in the reactor

Plasma electrolysis was conducted at some initial concentrations of tofu wastewater. We proved that plasma electrolysis can degrade COD at every initial concentrations of the wastewater, but the effectiveness of each processes would be different. As shown in Figure **??**, the best COD degradation was achieved at 2000 mg/L of the initial wastewater concentration. There were competitions between the initial reactant and the intermediate that was produced with active species [**?**]. Higher initial concentration of reactants made the competitions become more intense. Therefore, the percentage of COD degradation at 4000 mg/L and 7771.3 mg/L was lower than 2000 mg/L.

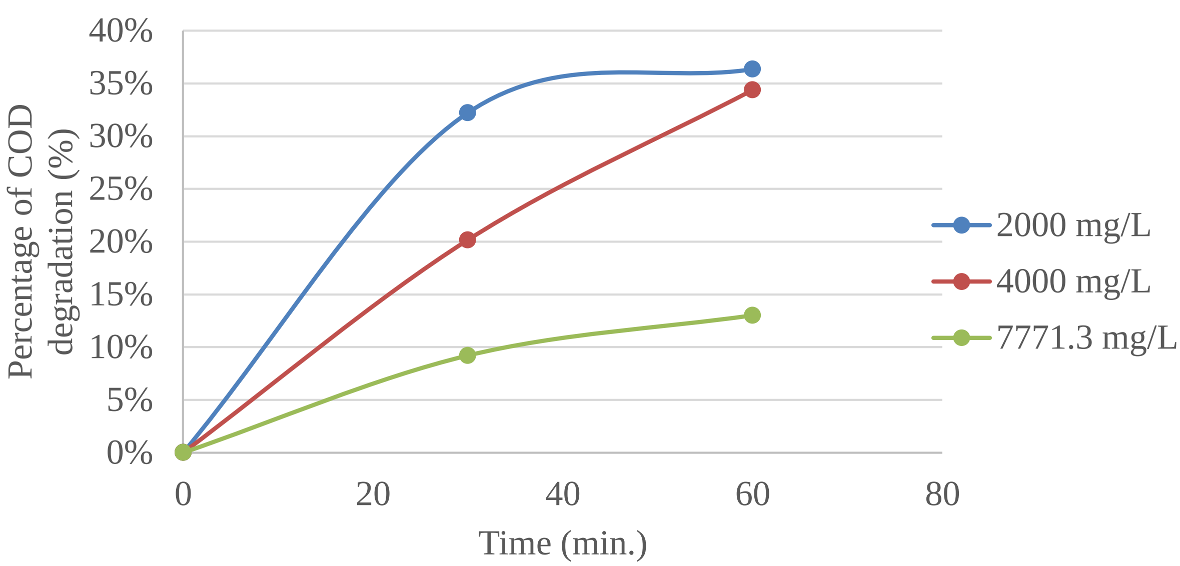

Fig. 6. The effect of initial concentration of tofu wastewater in plasma electrolysis

V. CONCLUSION

The combination of coagulation-flocculation and plasma electrolysis could degrade the pollution parameters in tofu wastewater effectively. Coagulation-flocculation with chitosan as the coagulant reduced COD and TSS, as much as 16.8% and 74.4%, respectively while plasma electrolysis as the advanced treatment decreased COD up to 36% at the initial concentration of tofu wastewater 2000 mg/L during 60-minute process.

Though the current research reduced the environmental damage caused by the wastewater with an effective treatment, yet further investigation in current domain is encouraged.

Declaration of Conflicting Interests

The authors make the declaration that no competing interests are present in this work.

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