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Research Article

Solar-Powered Electrocoagulation System for Tofu Wastewater Treatment and its Characteristic

Muryanto Muryanto^{1*}, Ajeng Arum Sari², Sunu Pertiwi², Danar Aji Prasetyo³, Sudarno³

¹Research Center for Chemistry, Indonesian Institute of Sciences, Kawasan PUSPIPTEK Serpong, Tangerang Selatan, Indonesia, 15314

²Research Unit for Clean Technology, Indonesian Institute of Sciences, Komplek LIPI Bandung Gedung 50, Jalan Cisitu Lama, Bandung 40135, Indonesia

³Department of Environmental Engineering, Faculty of Engineering, Diponegoro University, Jl. Prof. H. Sudarto, SH Tembalang, Semarang, Indonesia, 50275

* Corresponding author, e-mail: <u>moeryanto.mrt@gmail.com</u>

Abstract

This study aims to investigate the ability of solar-powered electrocoagulation for tofu wastewater, especially for reducing COD and TSS. This feasibility was compared with conventional electrocoagulation using electricity from the state electricity company. The study was conducted on a laboratory scale using a batch reactor electrocoagulation and aluminium electrode. The types of electrolytes used are sodium chloride and potassium chloride. The contact time is o, 2, 4, 6, and 8 hours. The results showed that removal of COD and TSS in tofu wastewater increases with a longer electrolysis time. During two hours of electrolysis time, the removal of COD and TSS were 25 and 53.85%, respectively. This process yielded the highest COD and TSS removal of 75 and 76.9%, respectively, at 6 hours. Pseudo-second order kinetics about COD removal, both in conventional and solar panel systems, is concluded. By adding NaCl electrolytes, the conductivity of wastewater was increased, and then the removal of COD and TSS was also increased. At the end of the electrolysis time (5 hours), the pH of wastewater was neutral. The results of sludge characterization using FTIR showed the presence of hydroxyl groups, amide compound, and aromatic compound. The process of using solar panels gives results slightly different from conventional electricity, but has advantages in terms of lower operating costs and environmental friendly.

Keywords: Electrocoagulation; electrolyte NaCl; solar panel; tofu wastewater

1. Introduction

At present, climate change and global warming have encouraged the development of energy technology. To minimize carbon emissions, several guidelines have been established. Traditional thermal power facilities are becoming increasingly unattractive. As a result, renewable energy such as hydro-energy, wind energy, solar energy, biomass energy, geothermal energy, tidal energy, and ocean thermal energy have seen fast developed in recent years. Solar energy is one of the most common renewable energies with some advantages. It has a relatively low conversion efficiency of 20%. Moreover, their feature provides a variety of output power with weather variations including partial shadowing circumstances (Hua et al., 2018). In terms of environmental principles, this ecological energy source is preferable used to tackle the environmental issues. Several studies have been reported that

solar energy appears to be promising for electrocoagulation (Patcharaprakiti et al. 2017; Nawalkar and Salkar., 2019; Oh et al., 2019). It is because electrocoagulation system may run on direct current (DC) and no need an inverter module, as is the case with most machinery (Phalakornkule et al., 2019).

Electrocoagulation is an electrochemical method in which the anode releases metal ions, such as aluminium and iron, as an active coagulant in the solution, and at the cathode, hydrogen gas was released through the electrolysis of water (Holt et al., 2002). If compare to the physical process such as screening, biology processes like activated sludge, and chemical processes using coagulation-flocculation for wastewater treatment, those techniques have some disadvantages (Liu et al., 2010). It require large space, took a long time, and need additional chemicals during the processes. In contrast, the electrocoagulation system has some advantages including require simple equipment, easy automation process and generate more significant destabilized particles due to the integration with the flocculation process caused by the turbulence generated from the gas production (Cañizares et al. 2006; El-Naas et al. 2009).

Electrocoagulation has been reported potential to treat various of wastewater such as municipal wastewater, water polluted with endocrine-disrupting chemicals, arsenic, manganese-phosphate and boron-containing wastewater and washing wastewater (Phalakornkule et al., 2019). Due to this ability, thus in this study, electrocoagulation system may possible to treat tofu wastewater. Tofu is one of the popular daily foods in some Asian countries, but its production generates a lot of wastewaters since the soaking process, soybean washing, filtering, tofu making, and equipment washing all use a lot of freshwaters. This wastewater contains a lot of organic pollutants with chemical oxygen demand (COD) and total suspended solids (TSS) values are 2,016 and 13,020 mg L-1, respectively (Prasetyo et al., 2018). Another paper reported that COD and TSS of the tofu industry also have relatively high values up to 6,785 mg L-1 and 3,100 mg L-1, respectively, and tend to be acid with a pH of around 4 (Bangun et al., 2013). This wastewater can disrupt water quality and a lower capacity of waters around the tofu enterprises. Some regulations about the wastewater quality standard, the maximum COD, TSS, and pH for tofu wastewater before discharge into the environment are 300 mg L-1, 200 mg L-1, and 6-9, respectively. Therefore, the appropriate treatment was needed to decrease COD and TSS concentration in tofu wastewater before being discharged to the environment or reused for another purpose. The Electrocoagulation system has been known can reduce COD and total solids up to 80.7% and 61.38% with an initial concentration of COD and total solids 332 and 984 mg L-1, respectively (Alex and Paul, 2015). Moreover, Karichappan et al. (2013) reported that an electrocoagulation system with a pH of 7, the current density of 15 mA, electrode distance of 5 cm and flow rate of 70 ml/min able to remove COD and TSS of rice mill wastewater. The initial concentration of COD and TSS were 2,200 and 768 mg/l, respectively can be removed up to 89%.

A solar-powered electrocoagulation system is a renewable technology. Previously, electrocoagulation system was carried out conventionally using a power source from State Electricity Company (PLN). Therefore, by considering the cost of conversion and the intermittency in time. This study aimed to compare the ability of electrocoagulation using conventional and solar panel systems to decrease COD and TSS concentration in the tofu wastewater. The effect of electrocoagulation on pH, temperature, and conductivity was investigated. For further analysis, the final product was also investigated by using FTIR. Solar energy is an unlimited energy source that can be converted into electricity, hence this study is expected to benefit areas with limited access to electrical power.

2. Material and Methods

2.1. Wastewater Samples

A sample of wastewater was collected from a tofu small enterprise community in South Tangerang, Banten, Indonesia. The tofu production reached 10 tons of soybean/month, producing as much as 20 m³ of wastewater. The wastewater produced condensed liquid separated from the tofu lump and containing high-degradable protein (Kaswinarni, 2007). The characteristics of wastewater used in

this study are listed in Table 1. After sampling, wastewater samples were collected in 20 litter's plastic containers for analysis and electrocoagulation treatments.

Table 1. Characteristic of tofu wastewater		
Characteristics	Values	
COD (mg/l)	2,016	
TSS (mg/l)	13,020	
Initial pH	3.56	
Temperature (°C)	27.4	

Reactor / Electrocoagulation Setup 2.2.

Both batch and the continuous experiment was executed on the laboratory scale. The electrocoagulation equipment was formed by three components: an electrocoagulation chamber, electrodes, and a monitoring instrument. The Electrocoagulation chamber was made of PVC that has 35 cm x 20 cm x 25 cm size. The electrodes used as anode and cathode were aluminium plates with 26.5 cm x 29.5 cm. Anode and cathode were arranged in a parallel system with the distance electrode 3.5 cm and use DC electric source. Figure 1 illustrated the scheme of equipment during the electrocoagulation process.

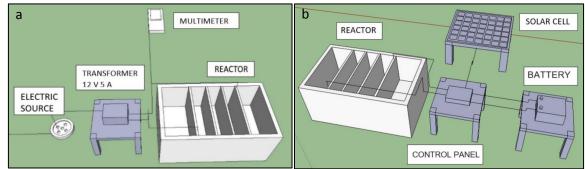


Figure 1. The electrocoagulation equipment. a) using conventional electricity, b) using solar panel

Methods of Analysis 2.3.

In the conventional system, electricity was applied with the transformer using 12V (Hamid, Purwono, and Oktiawan 2017) and a maximum of 5A. On the other hand, in the solar power system, two solar panels with each powered 50 W were connected to a battery of 12V, 45 Ah. Therefore, the study utilized varied contact time (0, 2, 4, 6, and 8 hours), different additional electrolytes (0.01M of technical KCl and NaCl), and electricity resources: from State Electricity Company mentioned as a conventional resource and from the solar panel.

Analytical Measurements 2.4.

The values of COD, TSS, temperature, pH, conductivity, and colour treated by the coagulation process were determined. COD parameter was identified by UV-vis spectrophotometer adapted from Indonesian National Standard (SNI) 06-6989.2-2009. Furthermore, TSS concentration was measured by the gravimetric method outlined from SNI o6-6989.3-2004. The investigation of temperature, pH, and conductivity was conducted by Benchtop Meters. Furthermore, Fourier transforms infrared spectroscopy (FTIR, Shimadzu, IR-prestige 21, Japan) was utilized to measure the functional group of the former and latter experience. In addition, scanning electron microscopy) (SEM Hitachi SU3500, Japan) was utilized for observing the sludge before and after the treatment.

3. **Results and Discussion**

3.1. The Effect of Contact Time and Additional Electrolyte on The Degradation of COD and TSS Concentration

The effects of supporting electrolytes, e.g. NaCl and KCl, were studied using conventional and solar panels as source power. Figures 2 and 3 show the variation of additional electrolytes to reduce COD and TSS using the conventional electricity and solar panel (Prasetyo et al., 2018). The results in Fig. 2 a and 2b show the same pattern, the COD reduction by conventional electricity was slightly higher than solar panel, but the value was not significant.

In early contact time of 2 h, the decline of TSS was obtained more than 50% in both conventional and solar-powered-electrocoagulation. Hereinafter, the TSS concentration does not change significantly until eight hours of operation. It is indicates that the optimum electrolysis time is 2 h. In contrast, COD concentration was decrease at each of contact time in both of conventional and solar-powered electocoagulation. This result was accordance with the research by Asselin et al. (2008) (Asselin et al., 2008). Over the first 2 h, the organic compound was removed by cathodic reduction—the organic matter deposited on cathode electrodes. The dispersed or suspended colloids of the organic compound were destabilized by neutralizing the potential energy of repulsion between charged particles, then flocculating by Al³⁺ from anodic dissolution (Asselin et al., 2008).

The electrocoagulation technique takes a long time to form flocks (Wang, Chou, and Kuo, 2009). The clumped flock adsorbed the dissolved organic compounds and then affected COD concentration in the wastewater. Following that, the flock was separated from the solution by hydrogen gas bubbles produced at the cathode electrodes. The significantly decreasing of COD and TSS after 2 h was also attributed to co-precipitation of organic matter with metallic hydroxides.

The destabilization process of organic compounds was due to the presence of dissolved Al^{3+} in wastewater. The flake of anode plate formed ion Al causing the electricity flow. This metal ion as contra ion has depleted the double-electric layer of organic pollutant to reducing the repulsion between particles. On the other hand, cathode, water (H₂O) was reduced and forming H₂ and OH⁻. Both anode and cathode reactions contained Aluminium hydroxide (Al(OH)₃), which served as a coagulant to destabilize colloid particles, thereby the capacity neutralization process particle causing particle adhesion with the opposite charge to promote the colloid aggregation and micro-suspension. Micro-flocculants were generated from the mixed colloid. As a result, it became larger and eventuallyy settled (Moussa et al., 2017).

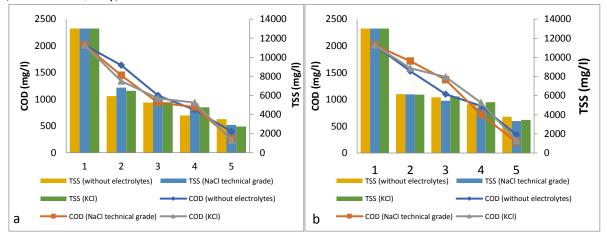


Figure 2. The decreasing of COD and TSS concentration during an electrocoagulation process using (a) solar panel electricity, (b) conventional electricity

The more formed hydrogen and hydroxide ion, the more complex system attached the pollutant declining COD. According to the first Faraday law, the charge total increases with the length of the electrolysis process. In other words, the wastewater containing pollutants was also accumulated in an

enormous amount. Likewise critical, the electrode had the maximum capacity to reduce organic wastewater. As a result, even if the electrodes were saturated to treat the wastewater, TSS could not be reduced any more, despite the contact time being increased.

3.2. The Effect of Contact Time and Additional Electrolyte on The Conductivity and Electricity Current

The addition of media conductivity and some treatments to the wastewater, which consist of the anion and cation, such as NaCl and KCl, Na₂SO₄, and KI, improved metal dissolution, decreasing the ohmic contact, including the cell voltage and energy consumption, and caused a surface phenomenon between the pollutant and metal hydroxide. Finally, the conductivity has an impact on the efficiency and power consumption of electocoagulation (Khandegar and Saroha, 2013; Chen, 2004). Ghosh et al. (2008) reported that 99.75 % of violet dye removal (initial dye concentration: 100 ppm) was obtained with the current density of 1,112.5 A.m⁻², solution conductivity of 1.61 Sm⁻¹, initial pH of 8.5, in 1 h operation time. By adding NaCl, the conductivity was increased about 11 times. The anode produced Cl₂ and OCl⁻ when chlorides were present in the solutions. The OCl is a strong oxidant that may oxidize organic molecules in a solution (Wong, 2002).

In addition, the electrocoagulation combined with additional electrolyte had higher efficiency than the solar panel or conventional electricity without it. The reason was caused by the chloride ion (Cl⁻) from additional electrolyte which prevent the passivity layer on the plate surface so that the electrocoagulation process worked properly (Liu et al. 2010).

Moreover, the conductivity affected the anode and cathode durability, as well as the electric current flowing in the wastewater (Zhang et al., 2013). The impact of contact time and the addition of supporting electrolytes on the conductivity of tofu wastewater was depicted in Figure 3a and 3b. The conductivity of wastewater increased while the additional electrolyte was added. As wastewater conductivity increased, so did the power of electrical flow (Chang, 2010); hence, electricity was effective in reducing pollution. At eighth hour, the wastewater conductivity was decrease in both of conventional electricity and solar panels. The decline caused by Chloride ion (Cl⁻) in the electrolyte changed to Cl₂ in the anode and composed ClO⁻ for pollutant removal (Zhang et al., 2013). Additionally, Cl⁻in the wastewater converted to active chloride that served as a desinfectant and reduced detrimental effect of CO_3^{2-} and SO_4^{2-} which releasing of Ca²⁺ and Mg²⁺ deposition on the oxide layer (Liu et al., 2010).

The solar panel gave the higher conductivity than conventional electricity. Although at the beginning of the process, the current was small, but in several hour the electric current generated by the solar panel is larger and more stable than conventional electricity. At high current, high pollutant removal was induced by two mechanisms: increasing the anodic dissolution of aluminium and the bubble production rate, resulting in a larger quantity of precipitate for pollutant removal (Shannag et al., 2012).

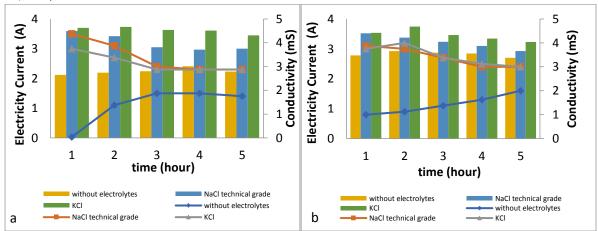


Figure 3. The effect of contact time and additional electrolyte to the conductivity of electrocoagulation process using (a) the solar panel, (b) the conventional electricity

3.3. The Effect of Contact Time and Additional Electrolytes on The Temperature and pH

Figure 6 showed the observation of the temperature of electrocoagulation using the conventional electricity and solar panel. The increasing of electricity current in the electrode plate improved the temperature of wastewater along the electrocoagulation process, as well as the solubility of aluminium (Perry and Green, 1997).

In this study, the temperature of wastewater through the conventional and solar-powered electrocoagulation processes were increased in eight hours. Based on results, both of these systems have a similar effect on pH and temperature (Figure 4a). According to Liu et al. (2010), the improvement of temperature encouraged the velocity of ions and particulates movement inside the liquid. These phenomena were followed by increasing the oxide layer destruction and current efficiency. However,

Sahu et al. (2014) reported that the high temperature decreasing the solubility of aluminium and current efficiency. Moreover, the temperature may affected the removal of COD during the electrocoagulation process. Yilmaz et al. (2008) state that when the temperature increased from 20° C to 60° C during the electrocoagulation process, the boron removal efficiency increased from 84 to 96% (Yilmaz et al., 2008).

Sahu et al. (2014) described the other crucial factor that affecting the electrocoagulation process is pH. pH can affect the wastewater conductivity, the solubility of an electrode, the form of hydroxide, and the zeta potential of colloid particles. In this study, the investigation of pH activity during the electrocoagulation process energized by solar panels and conventional electricity was described in Figure 4b.

Figure 4b showed that the pH of wastewater increased over 8 hours. It was contributed by the flocculates adsorbing the dissolved organic compound in tofu wastewater (Suwanto et al., 2017). In addition, the separation of water molecules to ion hydroxide impressed the alkaline characteristics of water. The pH increased more than previously because numerous ion hydroxides were produced during the electrocoagulation process.

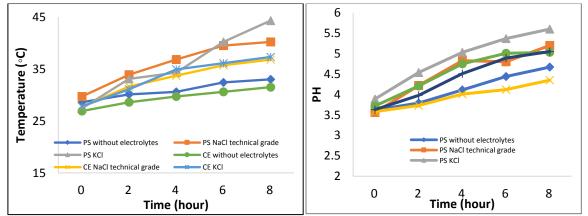


Figure 4. The effect of contact time and additional electrolyte to (a) the temperature (electricity source: solar panel); and (b) to pH

3.4. Kinetics Studies

COD reduction in electrocoagulation could be modelled as adsorption kinetic studies of conventional adsorption. The electrocoagulation process involves two steps: microparticle capture by hydrogen bubbles, followed by agglomeration on the free surface to form flocs (Benaissa et al., 2016). The pseudo-first-order and pseudo-second-order were modelled to investigate kinetic mechanisms of electrocoagulation within tofu wastewater treatment. As a result, the pseudo-first-order (eq 1 and 2) and

$$\frac{d_q}{d_t} = k_1(q_e - q) \tag{1}$$

pseudo-second equation (eq 3, and 4) are expressed as follows:

$$log(q_e - q_t) = log(q_e) - \frac{k_1}{2.303}t$$
(2)

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$\frac{d_q}{d_t} = k_2 (q_e - q)^2$	(3)
$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \left(\frac{1}{q_e}\right) t$	(4)

 q_e and q_t are the adsorption capacity at equilibrium, and at time t, k_1 is the rate constant of pseudofirst-order adsorption, and k_2 is the rate constant for second-order adsorption.

	State Electricity Company (PLN)			Solar cell		
	Without	NaCl	KCl	Without	NaCl	KCl
	electrolyte			electrolyte		
k	0.238394	0.190788	0.157071	0.192277	0.202236	0.14582
qe	1758.458	1694.659	1635.09	1690.469	2072.185	1840.285
R2	0.9789	0.953	0.9353	0.9966	0.8791	0.9345

 Table 2. Calculation of kinetic parameters and correlation coefficient in pseudo-second-order

The plot was found to be linear with a good correlation coefficient ($R^2=0.97$) for second-order kinetics, while pseudo-first-orders give small r2 values (data not shown). Thus, it can be concluded that the pseudo-second-order kinetic model describes better the adsorption of COD on aluminium hydroxide and fits experimental data better than the pseudo-first-order. The kinetic studies were in accordance with the electrocoagulation of potato chips wastewater treatment described in the study by Vasudevan et al. (2009).

3.5. Sludge Characteristic

3.5.1. FTIR (Fourier Transform Infrared Spectroscopy) Analysis

FTIR analysis was applied to characterize the functional group-containing the sample before and after coagulation process with additional NaCl, and the graphs of FTIR was expressed in Figure 5. The outcomes of functional group identification using FTIR in Figure 5 shows the molecule intensity values of tofu wastewater before treatment were lower than after the treatment process without additional electrolyte. The functional group's identification is presented in Table 3. According to Table 3, the sample shows a strong and broad at 3397 cm⁻¹; which corresponds to the hydroxyl groups (-OH). The peak at 1636 cm⁻¹ was presented the bend vibration of amine N-H which indicated the presence of amide compound. The vibration of C=C aromatic stretching groups are also shown at 1535 cm⁻¹; this indicates the presence of an aromatic compound in the sample. Aluminium hydroxide was found as a coagulant from aluminium plate disintegration after the sludge treatment (Gomes et al. 2007). Because of the addition of NaCl to the sludge following treatment, the chloro functional group was present.

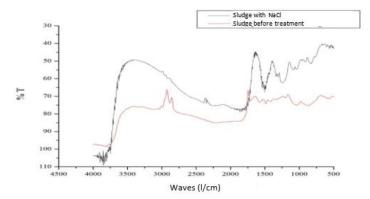


Figure 5. FTIR analysis of tofu wastewater treatment by product with the NaCl addition.

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	Before		After	
No	treatment	Functional groups	treatment	Functional groups
	(1/cm)		(1/cm)	
1	3397	-OH (Alcohol)	3375	-OH (Alcohol)
2	2925	C-H stretching	1634	N-H bending
3	1744	C=O stretching	1390	C-H bending
4	1636	N-H bending	1141	C-C-N (Amin)
5	1535	C = C aromatic	881	Al-O-H (Aluminum
				Hydroxide)
6	1369	C-H bending	779	C – Cl (Chloro)
7	1163	C-C-N (Amin)		

Table 3. The comparison of FTIR analysis on sludge before-after treatment using NaCl electrolyte

3.5.2. SEM Analysis

The surface of formed-latter sludge was visually examined using by SEM analysis to show the change in surface morphology on tofu sludge before and after electrocoagulation treatment. The investigation using 200, 2000, and 10,000 magnifications in each sample. Figure 6 and 7 showed the SEM before and after electrocoagulation processes.

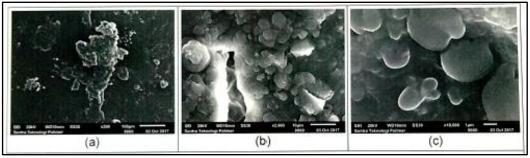


Figure 6. SEM analysis of tofu sludge before treatment (Magnification: (a). 200x (b). 2000x (c). 10000x)

SEM examinations defined and compared between untreated sludge and treated sludge which exhibit the difference in sludge morphology of layer surface. By increasing the size until 10000 times, before the electrocoagulation process, the layer surface morphology of tofu sludge was more compact than the later form. Furthermore, there are several large and delicate lumps attached to the surface of tofu sludge. Meanwhile, after electrocoagulation treatment, a substantial number of tiny granules spread out on the sludge surface in both with and without electrolyte addition.

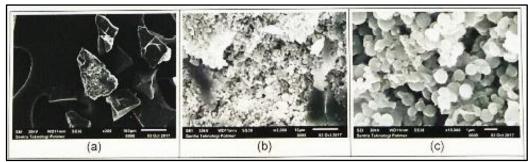


Figure 7. SEM (micrograph) analysis of tofu sludge after electrocoagulation treatment using NaCl addition (Magnification: (a) 200x (b) 2000x (c) 10000x)

4. Conclusion

The pH, COD, and TSS of tofu wastewater were 3.56, 2016 mg/L, 13020 mg/L, and 13020 mg/L, respectively. The sources of electricity through the electrocoagulation process were taken from conventional electricity produced by State Electricity Company and solar panels. The addition of NaCl as an electrolyte powered by the solar panel in eight hours was an effective method to increase the tofu wastewater conductivity. Furthermore, the temperature was then increased to 40-45°C, and the pH was progressively adjusted to neutral. Based on the FTIR investigation, aluminium hydroxide and chloro functional groups were found in the tofu sludge. The process of using solar panels gives results slightly different from conventional electricity, but has advantages in terms of lower operating costs and environmental friendly. To sum up, electrocoagulation energized by the solar panel as an electricity source became an alternative method to treat tofu wastewater, but the effectiveness solar panel in rainy season must be explored. Further, electrocoagulation system directly can be combined with simultaneous production of hydrogen for energy storage so that performance of this system can be enhanced.

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