



Coagulation and Filtration Methods on Tofu Wastewater Treatment

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ABSTRACT

The tofu industry is a small industry (home industry) that produces wastewater between 100-200 times the allowable limit and is usually discharged directly into water bodies, thus polluting the environment. This study aims to combine the coagulation method (stage 1) using Polyaluminium Chloride (PAC) with filtration (stage 2) on several variations of materials (quartz, activated carbon, and zeolite). The study was conducted with six replications. The comparison of waste quality (BOD, COD) was observed at each stage of the study. The SAS 9.4 was used for data analysis, including the application of the T-test and ANOVA. The study found that coagulation with PAC 690 mg/L reduced BOD by 51.7% and a dose of 765 mg/L by 61.1%. In the COD parameter, the reductions were 65.84% and 67.55%. In the second stage (filtration), the reduction in BOD was higher in activated carbon (79.33%) compared to zeolite (78.67%) and quartz (75.46%). Activated carbon also had the most COD reduction effect (73.22%). Although the statistical results showed significant differences in all doses and media, the use of 765 mg / L PAC and activated carbon filtration had the most effect on reducing BOD and COD of tofu industrial wastewater. This research can be used as an alternative in the physical processing of tofu industrial wastewater.

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Metode Koagulasi dan Filtrasi pada Pengolahan Limbah Cair Industri Tahu

ABSTRAK

Industri tahu dan tempe merupakan industri kecil (home industry) yang menghasilkan limbah antara 100-200 kali batas yang diijinkan dan biasanya langsung dibuang ke badan air, sehingga mencemari lingkungan. Penelitian bertujuan menggunakan metode koagulasi (tahap 1) dengan Polyaluminium Chloride (PAC), dan metode filtrasi (tahap 2) dengan tiga variasi bahan (kuarsa, karbon aktif, dan zeolit). Penelitian dilakukan dengan enam replikasi. Perbandingan kualitas limbah (BOD, COD) diamati pada setiap tahap penelitian. Perangkat SAS 9.4 digunakan untuk analisis data, termasuk penerapan uji T dan ANOVA. Penelitian mendapatkan, bahwa nilai BOD dan COD limbah segar industri tahu sebesar 1.813 mg/L dan 2.570 mg/L. Pada tahap pertama perlakuan (koagulasi dengan PAC 690 mg/L dan 765 mg/L) terjadi penurunan BOD sebesar 51,7%, dan 61,1%. Pada parameter COD, penurunan sebesar 65,84% dan 67,55%. Pada tahap kedua (filtrasi), penurunan BOD lebih tinggi pada karbon aktif (79,33%) dibandingkan dengan zeolit (78,67%) dan kuarsa (75,46%). Penurunan COD terbesar juga pada karbon aktif (73,22%). Walaupun hasil statistik menunjukkan perbedaan yang nyata pada semua variasi dosis dan media, namun penggunaan PAC dosis 765 mg/L dan filtrasi karbon aktif memberikan efek terbesar terhadap penurunan BOD dan COD limbah cair industri tahu. Hasil penelitian dapat digunakan sebagai alternatif dalam pengolahan fisika limbah cair industri tahu.

Kata kunci:

BOD
COD
Koagulasi
Filtrasi
Limbah-cair
Tahu

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INTRODUCTION

Water pollution has shown a serious problem in developing countries, including Indonesia. 75% of the total 57 rivers in Indonesia are heavily polluted (Dawud, Namara, Chayati, & Taqwa, 2016), caused by household waste (domestic) and untreated industrial waste (Rusydi, Suherman, & Sumawijaya, 2017). The tofu industry is an essential source of pollutants (Sudaryanto, 2006).

Tofu and tempe are Indonesian specialties made from soybeans (*Glycine spp.*) (Herlambang, 2002). Tofu and tempe are healthy, nutritious, and inexpensive foods, so they are favored by all levels of society (Alvina & Hamdani, 2019). The tofu industry is found in almost all provinces in Indonesia. Currently, Indonesia is the largest importer of soybeans in Asia (Nur Mahdi & Suharno, 2019). About 50% of soybeans are used for making tempe, 40% for tofu and the rest is in other products (Alvina & Hamdani, 2019). The largest soybean exporter to Indonesia is the United States, with a US\$ 4.8 billion in 2013 (Puspawati, 2017).

Most of the tofu industry is included in the small business group. Even though it contributes to society's economy, on the other hand, it harms the environment in the form of water pollution (Sudaryanto, 2006). The limited level of business capital causes the tofu industry cannot build a wastewater treatment plant, so it is discharged directly into the river. (Amanda, Marufi, & Moelyaningrum, 2019). Tofu wastewater was producing from the washing, boiling, and pressing processes of tofu. Each year, the potential wastewater of the tofu and tempe industry is an estimated 51 million tons of Biochemical Oxygen Demand (BOD) (Herlambang, 2002).

Tofu industry wastewater contains suspended (SS) and dissolved solids (DS) consisting of fat, protein, and cellulose, which will undergo physical, chemical, and biological changes to produce toxic substances if not appropriately treated (Amanda et al., 2019; Puspawati, 2017; Sudaryanto, 2006). Amino acids are the main component that causes pollution of water (Sudaryanto, 2006). Another study said that tofu industrial wastewater contains protein (40%-60%), carbohydrates (25% -50%), fat (10%), and other SS, which will produce toxic substances (Pradana, Suharno, & Apriansyah, 2018).

Several studies have reported varying quality of tofu wastewater. Puspawati's research results found (in ml/L units) Total SS=4,012.00, BOD=1,302.03, Chemical Oxygen Demand (COD)=4,188.27 (Puspawati, 2017). Alimsyah reported BOD=5,643-6,870 and COD=6,870-10,500 (Alimsyah & Damayanti, 2013). Bangun reports BOD=4,583, COD=7,050, TSS=4,743 (Ayu Ridaniati Bangun, Siti Aminah, Rudi Anas Hutahaean, & M. Yusuf Ritonga, 2013). Other studies found TSS=5,016, BOD=3,200, and COD= 4.200 (Amanda et al., 2019); TSS=1,850, BOD=1,130, COD=1,900 (Mardhia & Abdullah, 2018); and BOD=1.324, COD=6,698 (Herlambang, 2002). The results presented indicate that tofu wastewater is above the quality standard, TSS=200; BOD=150; COD=300 (Menteri Lingkungan Hidup, 2014). Therefore, tofu wastewater must be processed before disposed to the environment (Alimsyah & Damayanti, 2013; Ayu Ridaniati Bangun et al., 2013; Herlambang, 2002; Pradana et al., 2018; Puspawati, 2017; Sayow, Tilaar, & Naskah, 2020).

Wastewater treatment aims to reduce pollutants so that they are safely disposed to the environment. Pollutants in wastewater are mainly organic, inorganic, DS and SS. Environmental quality standards are the highest limits for the quality of wastewater so that it is safe to dispose into the environment. The primary environmental quality standards

are TSS, BOD, and COD (Menteri Lingkungan Hidup, 2014). The high content of pollutants in wastewater causes oxygen levels to decrease, thereby disrupting life in it (Pradana et al., 2018). Poor quality of water will impact reducing the number of biota in the water (Nangin, Langoy, & Katili, 2015; Yogafanny, 2015).

Many waste treatment methods have been developed: activated sludge, biological processes, electrocoagulation, filtration, coagulation, and flocculation—the industries with organic waste, the most widely used biological processes with aerobic and anaerobic. In the tofu industry, the technology that is often used is biologically an-aerobic because it is cheap. However, technology has disadvantages; it creates odors, produces methane gas, and an efficiency of 70-80%, so it still contains high pollutants (Herlambang, 2002; Said, 2000). Physical processing is alternative to reduce as much as possible solids content in the wastewater so that the biological process becomes most lighter.

This study analyzes the reduction of tofu wastewater parameters by physical treatment with two stages of processing—the first stage (coagulation) using two variations of Polyaluminium Chloride (PAC) dose. The second stage (filtration) used three variations of the filter. It is hoped that the research results can be a better alternative to tofu wastewater processing to reduce the impact of environmental pollution.

METHOD

Fresh wastewater is taken from the industrial tofu center in Gunung Sulah Village, Bandar Lampung City. PAC from a distributor of industrial chemicals; BOD and COD chemicals (H_2SO_4 , Ammonium Sulfate, $K_2Cr_2O_7$, and others) from Merck. The Messgerate model S6S six-spindle flocculator was used for the coagulation process. The research was conducted at the Tanjungkarang Health Polytechnic Laboratory.

The study was conducted in two stages to analyze the decrease in BOD and COD in tofu wastewater. The experiment was carried out with six replications. In the first stage, the coagulation process was carried out with PAC (dose 690 mg/L and 765 mg/L). PAC was added to fresh wastewater. The mixture was subjected to two minutes of rapid mixing at 180 rpm, followed by 25 minutes of mixing at 30 rpm and 30 minutes of settling. PAC dose 690 mg/L and 765 mg/L following Murwanto (2018).

The second stage is a filtration process with three variations: activated carbon, zeolite, and quartz. Each reactor is filled with different filtration media and set at a fixed rate. Substitution of the filter was carried out on each repetition to obtain the same conditions for each treatment. The filtration results are accommodated for testing the BOD and COD. BOD and COD were measured when fresh, after the coagulation process, and after filtration.

Measurement of BOD using the Winkler method. Two bottles (250 ml) of fresh wastewater were immediately taken, one bottle was measured for Dissolved Oxygen (DO day 0), and the other was stored at 4°C for the fifth day of measurement (DO₅). BOD is the difference of DO₀ and DO₅. BOD was measured at the end of each experimental stage. Measurement of COD levels using the Reflux method. Time of collection and measured together with DO₀ testing.

Data were analyzed using SAS (version 9.4) and were carried out in stages. Variable description with mean,

minimum-maximum, and percentage reduction in BOD and COD. The formula calculates the percentage reduction:

$$\% \text{ reduction} = \frac{(\text{Value before} - \text{Value after})}{\text{Value before}} \times 100\%$$

The T-test was applied to determine of effect the first stage treatment (coagulation) on the BOD and COD. In the second stage, ANOVA and Bonferroni tests were used to determine the effect of filter media variation on the BOD and COD.

RESULTS AND DISCUSSION

Table 1 shows the BOD and COD of fresh tofu waste after treatment stage I and stage II. The BOD of fresh wastewater was 1813.00 mg/L, after the coagulation process decreased to 790.00 mg/L (705.00-875.00), and after the stage II treatment became 417.03 (430.00-498.00). The percentage of reduction in BOD for fresh wastewater (Fig. 1) in stage I was 56.43% and 77.00% in stage II.

The COD of fresh wastewater was 2570.00 mg/L after the coagulation process decreased to 851.00 mg/L (824.00-878.00), and after the second stage, treatment became 830.19 mg/L (674.00-910.00). The percentage reduction of COD for fresh wastewater (Fig. 1) was 66.89% and 67.70%.

Table 1
BOD and COD measurement results

	BOD			COD		
	Fresh	Stage I	Stage II	Fresh	Stage I	Stage II
Mean (mg/L)	1813,00	790,00	417,03	2570,00	851,00	830,19
Minimum (mg/L)	1813,00	705,00	340,00	2570,00	824,00	674,00
Maximum (mg/L)	1813,00	875,00	498,00	2570,00	878,00	910,00
Reduction (%)	-	56,43	77,00	-	66,89	67,70

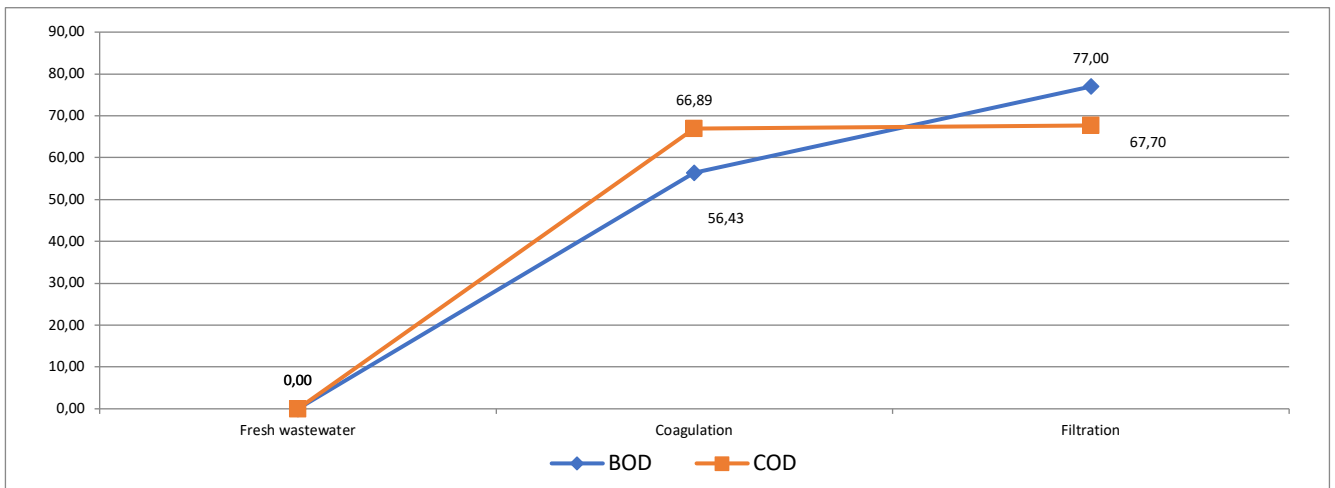


Figure 1. Percentage of reduction in BOD and COD based on treatment stages

The results showed that the coagulation process (Table 2) using a PAC dose of 690 mg/L could decrease the BOD from 1,813.00 mg/L to 875.00, or a reduction of 51.74%. Using PAC 765 mg/L can decrease the BOD from 1,813.00 mg/L to 705.00, or a reduction of 61.11%.

Reducing COD using a PAC 690 mg/L from 2,570.00 mg/L to 878.00 mg/L, or a reduction of 65.84%. Using a PAC 765 mg/L can decrease the COD from 2570.00 mg/L to 824.00 mg/L, or a reduction of 67.55%. Based on the coagulant dose, the PAC 790 mg/L had a more significant effect on reducing BOD.

Table 2
BOD and COD value after coagulation

	BOD			COD		
	Value (mg/L)	Reduction (%)	p-value	Value (mg/L)	Reduction (%)	p-value
Fresh	1,813.00	-		2,570.00	-	
690 mg/L	875.00	51.74	0.0001	878.00	65.84	0.0001
765 mg/L	705.00	61.11		824.00	67.55	

A T-test (alpha=0.05) was performed to see the difference in BOD and COD, before and after the coagulation process. The analysis results showed a significant difference in BOD

between before and after the coagulation process (p-value=0.001). The significant difference was also in the COD (p-value = 0.001).

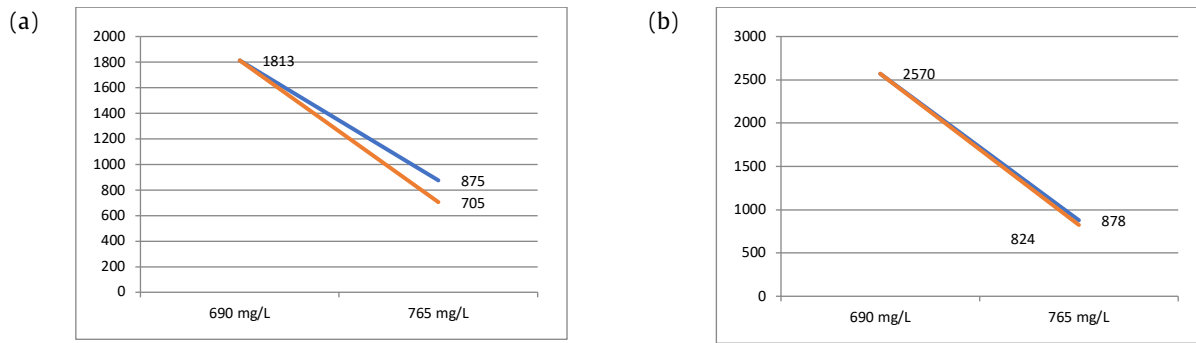


Figure 2. BOD (a) and COD (b) value, before and after the coagulation process

In stage two, the sample that has been coagulated with PAC is followed by a filtration process. Three types of media were used: quartz-sand, activated carbon, and zeolite (Table 3). In the sample group with a 690 mg/L coagulation dose, the highest average BOD using quartz (452.17 mg/L) and the lowest using activated carbon (405.50 mg/L). In the COD, the highest value was using quartz (896.33 mg/L), and the lowest was using activated carbon (834.00 mg/L). The first group used PAC 765 mg/L; the highest BOD with quartz (445.00 mg/L), and the lowest with activated carbon (374.83 mg/L). While the highest COD used quartz (866.33 mg/L) and the lowest used activated carbon (736.33 mg/L). The second stage treatment showed that the highest reduction in BOD

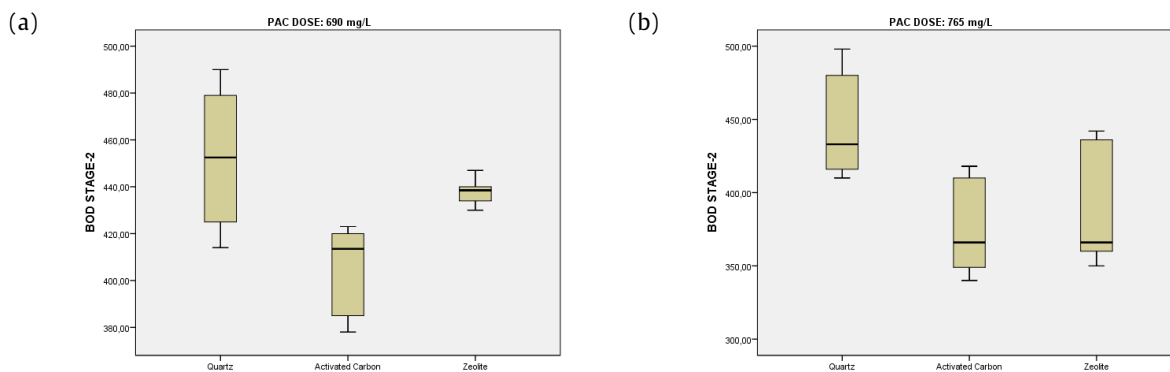
and COD used an activated carbon at a PAC dose of 690 mg/L or 765 mg/L.

ANOVA test was applied to determine the differences in BOD and COD after treatment based on variations of filter, both at the PAC 690 mg/L and 765 mg/L (Table 3). The sample group using 690 mg/L showed a significant difference in BOD based on the filter used ($p=0.007$). Likewise, the COD parameter also significant ($p=0.005$).

In the sample group using 765 mg/L, the BOD showed a significant difference based on the variation of media ($p=0.011$). Likewise, the COD parameter also significant ($p=0.015$).

Table 3
 BOD and COD values after filtration

PAC Dose	Parameters	Filters	n	Mean	SD	F	p-value		
690 mg/L	BOD	Quartz	6	452.17	32.74	7.009	0.007		
		Activated Carbon	6	405.50	19.11				
		Zeolite	6	438.00	5.76				
	COD	Quartz	6	862.00	27.42			7.864	0.005
		Activated Carbon	6	834.00	36.83				
		Zeolite	6	896.33	11.06				
765 mg/L	BOD	Quartz	6	445.00	35.79	6.221	0.011		
		Activated Carbon	6	374.83	33.25				
		Zeolite	6	386.67	41.18				
	COD	Quartz	6	866.33	27.41			5.589	0.015
		Activated Carbon	6	736.33	80.10				
		Zeolite	6	786.17	81.78				



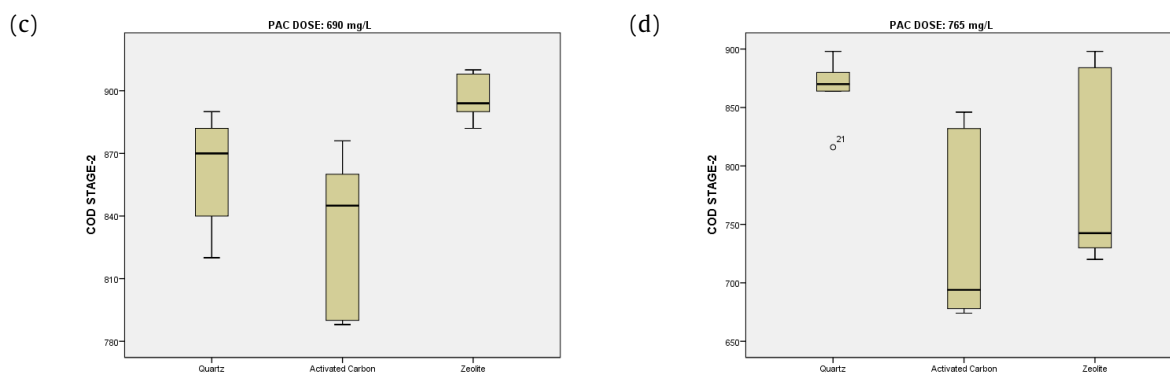


Figure 3. BOD and COD values after filtration with various media at a PAC dose of 690 mg/L (a) (c), and 765 mg/L (b) (d).

Table 4
 Bonferroni test ($\alpha=0,05$)

PAC Dose	Parameters	Filter (p-value)			
		Quartz	Activated Carbon	Zeolite	
690 mg/L	<i>BOD/COD</i>	Quartz	-	0.007*/0.287	0.855/0.137
		Activated Carbon	0.007*/0.287	-	0.068/0.004*
		Zeolite	0.855/0.137	0.068/0.004*	-
765 mg/L	<i>BOD/COD</i>	Quartz	-	0.015/0.014	0.046*/0.177
		Activated Carbon	0.015*/0.014*	-	0.015*/0.670
		Zeolite	0.046*/0.177	0.015*/0.670	-

* signifikan ($\alpha < 0,05$)

Tofu is a food made from the main ingredient of soybeans (*Glycine spp.*), a simple manufacturing process. The principle of making tofu is to extract protein, then coagulate it with the help of CH_3COOH or $\text{CaSO}_4 \cdot \text{nH}_2\text{O}$ (Herlambang, 2002). Tofu industrial wastewater comes from the washing, boiling, and pressing processes of tofu, containing protein (40%-60%), carbohydrates (25%-0%), fat (10%), SS, and DS, which will produce toxic substances if not processed (Amanda et al., 2019; Pradana et al., 2018; Puspawati, 2017; Sudaryanto, 2006).

Generally, tofu waste treatment is carried out using anaerobic biological systems (Herlambang, 2002). The anaerobic wastewater treatment process is a metabolism without oxygen and is carried out by anaerobic bacteria, usually used for wastewater with a high organic matter (Rahadi, Wirosoedarmo, & Harera, 2018). However, this processing method produces methane gas (CH_4) which causes an odor and an efficiency of 70-80%, so that it still contains high pollutants (Herlambang, 2002; Said, 2000).

The results showed that the BOD of tofu industry wastewater was 1,813.00 mg/L, and COD was 2,570.00 mg/L; it is far from the quality standard (BOD=150 mg/L; COD=300 mg/L) (Menteri Lingkungan Hidup, 2014). The high content of pollutants can cause pollution of water (Sudaryanto, 2006). So it must be processed before disposing to the environment (Alimsyah & Damayanti, 2013; Ayu Ridaniati Bangun et al., 2013; Herlambang, 2002; Pradana et al., 2018; Puspawati, 2017; Sayow et al., 2020).

In this research, for the first stage treatment (coagulation) using a PAC 690 mg/L, there was a decrease in BOD from 1813.00 mg/L to 875.00 mg/L (51.74%), and COD from 2570, 00 mg/L becomes 878.00 mg/L (65.84%). Using PAC 765 mg/L, BOD decreased to 705.00 mg/L (61.11%), and COD to 824.00 mg/L (67.55%). These results indicate that the coagulation process with PAC can reduce BOD and COD by

more than 50%. Using a dose of 765 mg/L can reduce it to more than 61%.

The coagulation is the technology most widely applied in the world as a vital step in removing colloid particles, natural organic matter, microorganisms, and inorganic ions from untreated water (Ahmad & Danish, 2018; Kakoi, Kaluli, Ndiba, & Thiong'o, 2016; Kristianto, 2017; Maurya & Daverey, 2018; Muthuraman & Sasikala, 2014; Sillanpää, Ncibi, Matilainen, & Vepsäläinen, 2018; Tripathy & De, 2006). Coagulants are essential ingredients in the coagulation-flocculation process, which refers to the agglomeration process of colloid particles with an average size of 5-200 nm and small SS in water (Carolin, Kumar, Saravanan, Joshiba, & Naushad, 2017; Debora Perucó Theodoro, Felipe Lenz, Fiori Zara, & Bergamasco, 2013; Fu, Meng, Lu, Jian, & Di, 2019; Hakizimana et al., 2017; Kristianto, 2017; O'Connell, Birkinshaw, & O'Dwyer, 2008; Salehizadeh, Yan, & Farnood, 2018; Senthil Kumar et al., 2019; Shen, Gao, Guo, & Yue, 2019; Sillanpää et al., 2018).

There are four mechanisms in coagulation: double layer compression, polymer bridging, neutralization, and coagulation sweep (Kristianto, 2017). Double-layer compression is caused by high electrolyte concentrations in solution, thereby reducing the repulsive force of colloidal particles. Bridging polymer usually occurs when long-chain polymers are adsorbed on particles and leave the coagulant polymer segments to bridge the particles, forming strong lumps. Charge neutralization occurs when coagulant polymers with opposite charges are absorbed on the surface of the particles. So that will be neutralizing the charge of colloidal particles. Coagulation sweep occurs when metal coagulants are added in doses that are much higher than the solubility of amorphous hydroxides, resulting in precipitation (Duan & Gregory, 2003; Kristianto, 2017).

Of the four mechanisms, bridging is the best mechanism (Kristianto, 2017). However, it takes the correct dose. At higher doses, colloid particles are stabilized due to steric repulsion from the polymer covering the particles. Instead, there are not enough polymer chains at low doses to form a bridging process (Kristianto, 2017; Tripathy & De, 2006). In this study, a dose of 765 mg/L could form a bridging process and was stable, making it practical for use in tofu wastewater treatment. The correct coagulant dose will increase turbidity significantly, be avowed effective if it can reduce solids by at least 50% (Ayu Ridaniati Bangun et al., 2013).

In this study, the coagulant used is PAC, a synthetic polymer that is easier to hydrolyze, has a long molecular chain, and a large electric charge. The advantages of PAC in wastewater treatment are forming flocks quickly, produce less sludge, and produce small aluminum residues (Ignasius, 2014). The analysis results (Table 2) show a significant difference in BOD and COD between before and after the coagulation process (p -value=0.001; p -value=0.001). It shows the effect of the coagulation process on the decrease in BOD and COD. This study consistent with the previous studies, which concluded that the higher the dose, the greater the suspended solids would be removed (Ayu Ridaniati Bangun et al., 2013; Murwanto, 2018; Rahmah & Mulasari, 2016).

Three types of media are used in the filtration: quartz, activated carbon, and zeolite (Table 3). The results showed the highest decrease BOD with activated carbon, from 705.00 mg/L to 374.83 mg/L; or 46.83% compared to the results of phase I, and 79.33% compared the fresh wastewater. Likewise, for COD, the highest decrease was also using activated carbon, from 824.00 mg/L to 736.33 mg/L; or 10.64% compared to stage I and 71.35% compared to fresh wastewater COD. Statistically, activated carbon media had the most significant effect on reducing BOD and COD (Table 3).

This study is consistent with the previous study, which concluded that activated carbon was effectively used in tofu wastewater treatment, reducing BOD from 333.2 mg/L to 294 mg/L, and COD from 666.4 mg/L to 588 mg/L (Budiman & Amirsan, 2015); COD from 3,200 mg/L (Alimsyah & Damayanti, 2013). These results are also consistent with several other studies (Astuti, Wisaksono, & Nurwini, 2007; Larasati, Andita, Susanawati, Liliya, & Suharto, 2015; Lempang, 2014; Murwanto, 2018; Nurlina, Zahara, Gusrizal, & Kartika, 2015; Purnawan, Martini, & Aidah, 2014; Ronny & Syam, 2018; Sutapa, 2014).

In addition to filtration, there is also an adsorption process of pollutant molecules on the activated carbon surface. So, the concentration will decrease. Adsorption occurs because there is a force field on the adsorbent surface that attracts the adsorbate molecules, forming a thin layer on the surface of activated carbon. (Nurlina et al., 2015). The coagulation process with PAC 765 mg/L and followed by filtration using activated carbon reduced BOD to 374.83 mg/L and COD by 736.33 mg/L. This result is equivalent to a decrease of 79.33% and 71.35% compared to the initial conditions (fresh wastewater). Although the final results have not met the specified quality standards, it has proven that the coagulation and filtration methods can reduce BOD and COD by above 60%.

CONCLUSIONS AND SUGGESTIONS

The results showed that the BOD and COD of tofu

industrial wastewater were 1,813.00 mg/L and 2,570.00 mg/L, far exceeding the required quality standards. The coagulation process with PAC affected reducing BOD and COD by 61.11% and 67.55%. In the filtration process, activated carbon significantly decreased waste parameters, reaching 79.33% for BOD and 71.35% for COD. So that the final BOD value is 374.83 mg/L, and COD is 736.33 mg/L. Although the final results of the treatment have not met the specified quality standards, research has proven that physical processing with coagulation and filtration methods can reduce BOD and COD in tofu wastewater by above 60%. The results of this research can be used as an alternative in the physical processing of tofu industrial wastewater to improve the performance of biological processing at a later stage.

Conflict of Interest

The authors declare there is no conflict of interest.

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