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Ability of Ceramic Tiles Waste as a Pre-treatment for Laundry Wastewater

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Abstract. The discharge of laundry of laundry wastewater (LWW) into rivers contaminates the water and exposes it to harmful chemicals present in detergents and fabric softeners. This draws attention to the need to implement treatment for LWW. This study focused on determining the ability of ceramic tiles to remove total phosphorus (TP) and chemical oxygen demand (COD) from commercial LWW. The coarse aggregate of ceramic tile waste (CTW) was used as the adsorbent. The effectiveness of CTW as an adsorbent to remove TP and COD in LLW was determined by using different adsorbent dosages, contact times, and shaking speeds in a batch experiment. LWW samples were collected from the discharged point of commercial laundry shop. The results revealed that the highest TP removal was 71% with a dosage of 6 g/100 ml ceramic adsorbent, a contact time of 90 minutes, and a shaking speed of 100 rpm. Meanwhile, the highest removal of COD was 80% at a dosage of 6 g/100 mL of ceramic adsorbent, a contact time of 90 minutes, and a shaking speed of 300 rpm. The optimal value of removal for COD 60 mg/L and TP is 1.79 mg/L while pH value is 7.13. Thus, it can be concluded that the CTW aggregate as an adsorbent was effective in reducing TP and COD from LWW.

1. Introduction

Commercial laundries are among the service industries that consume a lot of water and consequently produce a large amount of wastewater. LWW exerts detrimental effects on both the environment and human health if discharge untreated. Studies had found that LWW from washing machines has significant amounts of nitrogen, phosphate, heavy metals, linear alkylbenzene sulfonates (LAS), volatile organic acids, and alcoholic chemicals, all of which can have harmful effects on the ecosystem and biodiversity [1,2,3]. Eutrophication of water bodies is caused by phosphorus content that originated from various sources of wastewater such as LWW. Thus, various techniques such as biological processes, physicochemical processes, filtration, and adsorption are used to treat or reuse laundry wastewater to remove or minimize these pollutants [4]. Among these techniques, adsorption is a feasible and relatively inexpensive approach to treat laundry wastewater [5,6]. Adsorption is a simple method to perform, has a wide pH range and high metal binding capacity [7]. The use of adsorption processes to remove parameters has attracted much attention in recent years [8].

Currently, the use of waste as adsorbent material has become a research edge which resulted from the quest of finding a sustainable or green solution that applies the cradle to grave concept. Ceramic waste is a type of adsorbent that offers many advantages. Clay-based ceramics have high ion exchange potential and can absorb some anions and cations, which are exchanged for other anions and cations in aqueous solution [9]. Ceramics can remove impurities, which greatly facilitates the adsorption and absorption processes. Porosity is a fundamental requirement for ceramic substrates to perform well in solid-gas reactions at high temperatures; it should be as stable as possible throughout use to minimize compaction even after multiple treatments at high temperatures [9]. Ceramics have a pore diameter of less than 30 μm and are chemically resistant to weak acids and bases [10].

According to Wen et al. [11], the ceramic was very effective in reducing TSS and ammoniacal nitrogen (NH_4-N) in combined effluents, with up to 100% and 95% effectiveness, respectively. While, according to Malapane and Hackett [12], ceramics have 83% to 99% effectiveness in reducing turbidity in domestic wastewater. Since the coarse aggregate from ceramic tile waste had a smaller size, it performed better in the removal parameters of wastewater [3]. This study evaluates the removal efficiency of each parameter by adsorption processes using coarse aggregate from ceramic tile waste with different adsorbent dosages, contact times, and shaking speeds.

2. Materials and Methods

2.1 Preparation of CTW

The ceramic tile wastes were collected from Gian Hong Tiling & Trading Sdn. Bhd. Parit Raja, Batu Pahat, Johor. The waste was brought to the laboratory in a tightly sealed plastic bag. The ceramic tiles were crushed and pulverized using a jaw crusher machine at the Construction Engineering Laboratory, UTHM. A sieve was used to separate the aggregates and the powder. The size of the ceramic waste was 2 mm (for batch study) and the powder size was 63 μm (for characteristics analysis) at the Geotechnical Engineering Laboratory of FKAAB, UTHM. Final products were kept in plastic container for further use.

2.2 Fourier Transform Infrared (FTIR) spectroscopy

FTIR was used to identify surface functional group of CTW sample. The Agilent Technologies Cary 630 FTIR is used to examine a 63μ sample of raw ceramic tile waste.

2.3 Filed Emission Scanning Microscopy (FE-SEM)

FE-SEM was conducted to determine the surface morphology of the microstructure image CTW materials. The CTW samples were mounted on the copper stub using adhesive carbon tape. The analysis was conducted at Microelectronics and Nano Technology-Shamsuddin Research Centre (MINT-SRC) laboratory, UTHM.

2.4 LLW Sampling

Samples were collected from the effluent point (figure 1) of LLW at Dobi Al-Hijrah, at Parit Raja, Batu Pahat, Johor. The sampling and storage were carried out according to APHA standard method. Samples of LLW was collected manually by grab sampling and immediately transferred into a cool room of 4^0C at wastewater laboratory, UTHM. The analysis of LWW characteristics were measured according to standard method (table 1).

2.5 Batch Method Experiment

In the batch method, orbital shakers were used to mix, blend, or agitate samples and substances in a flask by shaking. It contains an oscillating plate on which the flasks are placed. The batch method, in which the dosage of adsorbent in the solution, contact time, and shaking speed were varied, was used in all batch tests. In this method, different adsorbent dosages, contact times and shaking speeds were used to study the results of contact time as a function of shaking speed, using different total adsorbent dosages such as 3 g, 6 g and 9 g. Figure 2 shows the diagram of laundry wastewater during the shaking process while the process was used in the batch experiments with the shaking set.

Figure 1. The discharge point of collecting laundry wastewater

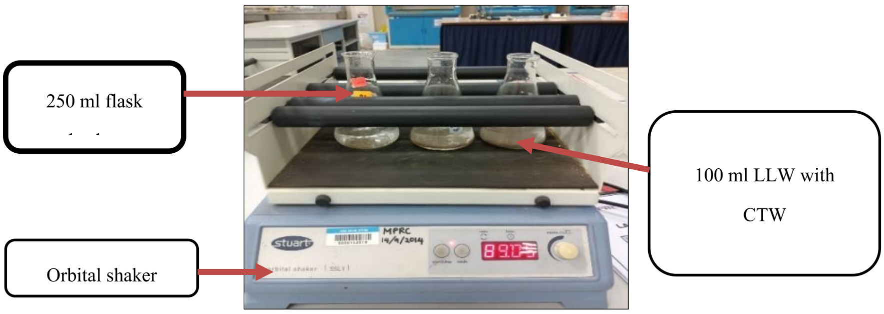


Figure 2. Laundry wastewater during shaking process in a batch method

2.6 Optimization process using RSM

Respond surface methodology (RSM) was used for optimizing the best conditions for pH, COD, and TP removal from LWW. To account for all potential combinations of factor values, 20 runs were completed. Adsorbent dosage (3–11g), contact duration (60–120min), and shaking speed (100–300rpm) were chosen as independent variables to explore the whole range of variables that could affect the reduction of pH, COD, and TP removal. For each run, a fixed volume of the LLW sample (100 mL) was mixed with different CTW dose and mixed for different times according to the expert designed program suggestions. To measure the efficiency of the CTW the percentage reduction of TP and COD were calculated by using Eq. (1).

$$\% \, removal = \frac{(Co - Cr)}{Co} \text{x } 100 \tag{1}$$

Where: C_0 is the initial concentration (mg/L), C_r represent the final concentration (mg/L) in the filtrate.

2.7 Statistical Analysis
The interaction between independent factors as well as first, second order model was subjected for statistical analysis using ANOVA with 95% of confidential level. The interactions and effects of the independent factors were considered as significant at $p < 0.05$.

3. Result and Discussions

3.1 Characteristic of Laundry Wastewater (LLW)
Raw laundry effluent results were evaluated and compared to previous studies and the Environmental Quality Act (EQA) of 1974 in the Environmental Quality (Sewage) Regulations, Second Schedule Regulation 7 for Acceptable Conditions of Sewage Discharge of Standards A and B. For effluent discharge standards A and B, Table 1 lists the parameters that can be used to distinguish the effluent discharge within the standard compared to the literature review from the previous study and the Environmental Quality Act of 1974.

Table 1. Characteristic of laundry wastewater compared with EQA 1974 and previous study

Parameter	Unit	Laundry wastewater Dobi-Al-Hijrah	EQA, 1974 Standard A	EQA, 1974 Standard B	Previous study
pH	mg/L	9.80	6.0 – 9.0	5.5 – 9.0	3.3 – 8.3 [13]
COD	mg/L	127	120	200	220-280 [14]
TP	mg/L	4.38	5.0	10.0	4-27.6 [15]

Table 1 shows that the pH of the wastewater sample from Dobi Al-Hijrah laundry was 9.80, which indicates that the sample was alkaline. The results show that the laundry wastewater from Dobi Al Hijrah is not acceptable because the pH of the wastewater exceeds the EQA 1974 quality standard, which requires a value between 6.0 and 9.0 for standards A and B, respectively. It was also higher than the pH of the previous study, which was still within the acceptable range of the standard. In addition, the volume of COD in the laundry effluent of Dobi Al-Hijrah was 127 mg/L, which was higher than the permissible value of 120 mg/L of standard A, but lower than the permissible value of 200 mg/L of standard B. Therefore, the effluents from Dobi Al-Hijrah can be discharged to other inland waters or Malaysian waters, but not to inland waters within catchments (EQA, 1974). Moreover, the COD values of laundry effluents from Dobi Al-Hijrah are lower compared to previous studies. The value of total phosphorus (TP) in the laundry wastewater is 4.38 mg/L, which is lower than the acceptable value of standard A or B compared to EQA 1974. However, the amount of TP in the laundry wastewater is quite close to the acceptable value of 5.0 mg/L of standard A. Compared to previous studies, the concentration of TP in the laundry's wastewater exhibits a noticeable increase.

3.2 Characteristic of the CTW

3.2.1 FTIR Analysis.
Samples of ceramic tile waste examined by infrared spectroscopy contained functional groups, C=O, C-O=H, C=C group vibrations, C-N amines, and C-H aromatics. The functional group of NH_2- and NH_3-amine salts was also included in the FTIR test results. The FTIR test results on the ceramic tile waste samples showed that the peak of wavelength is at 1006.38 with intensity of 100 and followed with 775.29, 693.28, 674.65, and 663.47 peak value of wavelength. The major functional group on the CTW were C=C, C=O, C-O=H and were responsible for the removal of the physic-chemical parameters in LLW [3]. The maximum intensity values in the FTIR spectrum are necessary for the identification of the constituents of the ceramic samples [16,17]. The FTIR spectra of the tile samples in the range of OH stretching vibration of hydroxide were more broadened when a higher firing temperature was used in the production of the tile bodies [16,17]. It is obvious that the FTIR spectrum is broadened between 3328 and 3250 cm-1. Therefore, due to the high intensity and broad spectrum for adsorption of

parameters in laundry wastewater, ceramic tile waste is intended as a good adsorbent. The peak value of IR spectrum is shown in figure 3.

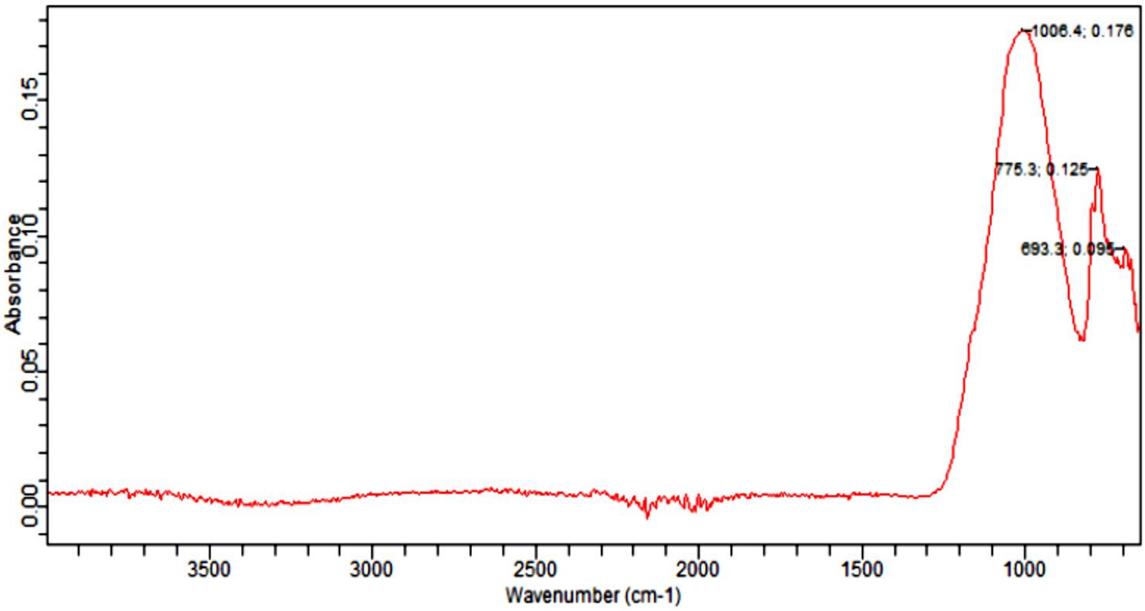


Figure 3. FTIR spectra of CTW

3.2.2 FESEM-EDS Analysis.

Magnifications of 1000 X and 2000 X were used at SEM to analyze the surface morphology of the ceramic aggregate at a particle size of 200 mm. The processed CTW has an irregular and uneven outer surface with crater-like pores that promote the accumulation of toxic metal ions, as shown in the microscopic image SEM. CTW exhibits rough and large voids. Energy dispersive X-ray spectroscopy at a radiation of 20.00 kV (figure 4) was used in conjunction with SEM to further determine the elemental composition of the ceramic aggregate. Analysis of SEM revealed that the CTW consists of the elements oxygen (O), silicon (Si), aluminum (Al), potassium (K), iron (Fe), sodium (Na), and calcium (Ca). The highest percentages of composition are contained in O and Si. These materials have a high percentage of parameter removal and absorption [18].

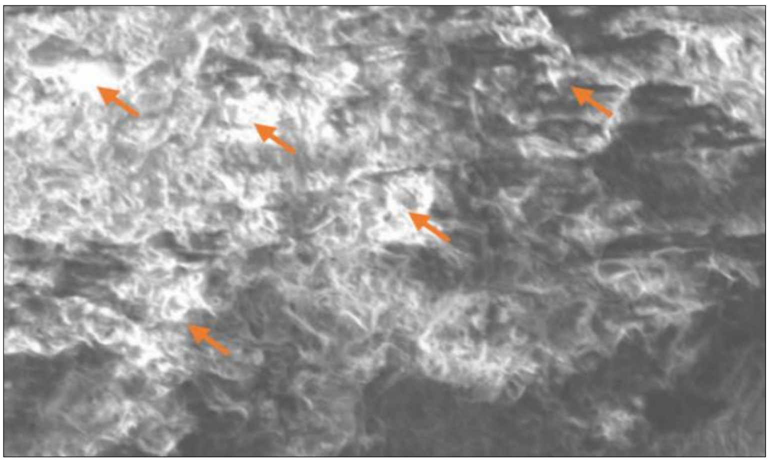


Figure 4. FE-SEM of CTW

3.3 Response Surface Methodology (RSM) Optimization

The efficiency of CTW in removing pH, COD, and TP from the LLW are presented in Table 2 and Table 3. The results revealed that the highest TP removal was 71% with a dosage of 6 g/100 ml ceramic adsorbent, a contact time of 90 minutes, and a shaking speed of 100 rpm. Meanwhile, the highest removal of COD was 80% at a dosage of 6 g/100 mL of ceramic adsorbent, a contact time of 90 minutes, and a shaking speed of 300 rpm. These findings revealed the potential of CTW in removing pH, COD and TP from the LLW. The ANOVA analysis revealed that the CTW dosage, contact time, and shaking speed play important role in removing pH, COD and TP (Table 2).

The linear and quadratic equation regression coefficients are presented in the Eqs 2–4.

$$Y_{pH} = 7.18 + 0.0272A + 0.1485B - 0.1778C - 0.0113AB - 0.0687AC - 0.2412BC - 0.0463A^2 + 0.3232B^2 - 0.0587C^2 \quad (2)$$

$$Y_{COD} = 42.55 - 2.03A + 1.73B - 5.15C \quad (3)$$

$$Y_{TP} = 1.64 + 0.1040A - 0.0348B + 0.0763C - 0.0438AB + 0.0112AC + 0.0387BC - 0.0041A^2 - 0.0766B^2 - 0.0500C^2 \quad (4)$$

where X1 (Adsorbent Dosage), X2 (Contact Time), X3 (Shaking Speed), Y1 pH), Y2 (COD), Y3 (TP)

Table 2. Central composite design arrangement and response for removing pH, COD and TP from LLW using CTW

Run	X1 Adsorbent Dosage (Gram)	X2 Contact Time (min)	X3 Shaking Speed (rpm)	Y1 pH	Y2 COD removal (%)	Y3 TP removal (%)
1	3	60	300	7.22	63.78	66.21
2	3	60	100	6.58	59.84	69.43
3	3	120	300	7.38	66.93	67.12
4	6	120	200	8.09	64.57	69.63
5	3	120	100	7.79	52.76	68.04
6	9	60	300	7.13	65.35	60.73
7	6	90	200	7.2	63.78	64.84
8	6	90	200	7.2	63.78	64.84
9	6	90	100	7.89	66.14	70.55
10	6	39	200	8.46	80.21	67.12
11	9	60	100	6.85	60.63	59.13
12	9	90	200	7.26	76.38	61.19
13	6	90	300	6.5	80.31	62.79
14	3	90	200	7.2	75.59	66.21
15	9	120	100	7.93	64.57	67.58
16	6	90	200	7.16	62.99	61.19
17	6	90	200	7.16	62.99	61.19
18	6	90	200	7.16	62.99	61.19
19	6	90	200	7.16	62.99	61.19
20	9	120	300	7.33	73.23	59.59

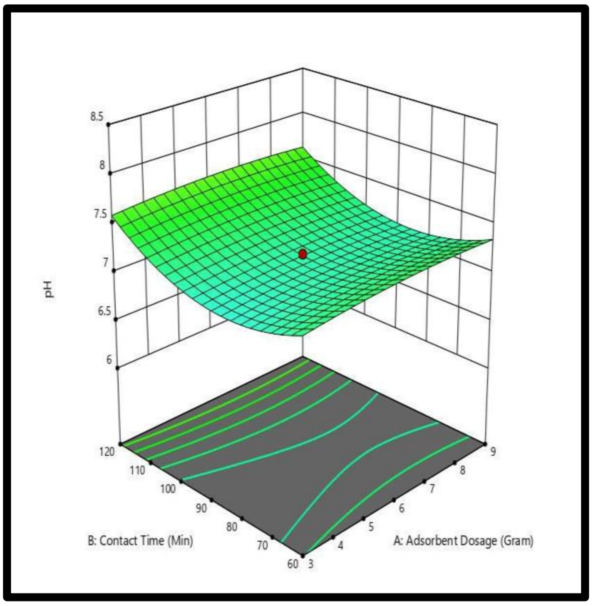
Based on the results of ANOVA analysis, it was noted that the models of pH, COD were not significant, while for TP was significant with a p-value of 0.1207, 0.1207 and 0.0216 respectively. The ANOVA analysis showed that adsorbent dosage has a linear effect on the pH, COD and TP, while, quadratic effect on pH and COD with a p-value <0.05. Furthermore, the ANOVA analysis displayed an interaction between X1 and X3 affected the responses of Y3 significantly (p-value<0.05). Moreover, the model of Y1, Y2 and Y3 can be considered as a significant with a confidence level more than 90%.

Table 3. Show significant of the quadratic model for removing pH, COD and TP from LLW using CTW

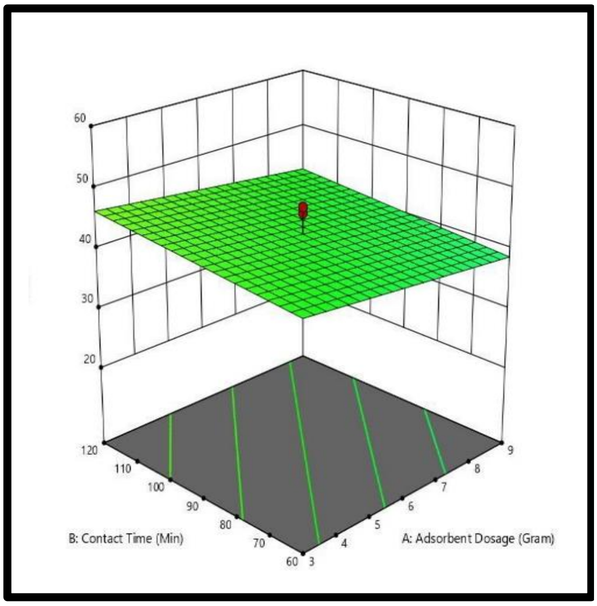
	F-value			P value		
	pH	COD	TP	pH	COD	TP
Model	2.18	2.26	3.95	0.1207	0.1207	0.0216
X_1 (Adsorbent Dosage)	0.0670	0.8295	13.69	0.8010	0.3759	0.0041
X_2 (Contact Time)	2.00	0.6047	1.53	0.1873	0.4481	0.2441
X_3 (Shaking Speed)	2.87	5.35	7.37	0.1210	0.0344	0.0218
X_1X_2	0.0067	1.42	1.42	0.9362	0.2610	0.2610
X_1X_3	0.2516	0.0939	0.0939	0.6268	0.7656	0.7656
X_2X_3	3.10	1.11	1.11	0.1089	0.3161	0.3161
X_1^2	0.2516	0.0222	0.0222	0.6600	0.8845	0.8845
X_2^2	10.02	7.83	7.83	0.0101	0.0189	0.0189
X_3^2	0.3300	3.35	3.35	0.5784	0.0973	0.0973

The graph 3D produced by quadratic equation showed interaction between the independent factors as shown in Figure 5. RSM analysis performed 3D graphical for each removal metals to determine the interactions between the variable factors and affect the percentage removal of pH, COD and TP. According this study 3D curve plot represented ceramic composite bead versus concentration as important factors affected to removal of pH, COD and TP in LLW. According to the Figure 5(a), the interaction between adsorbent dosage and CTW has been recorded the optimum removal of pH (7.13). Regarding the effect of interaction between contact time and CTW showed the optimum of removal COD content 60 mg/L (Figure 5(b)). The interaction between adsorbent dosage and CTW has been recorded the optimum of removal TP 1.79 mg/L (Figure 5(c)).

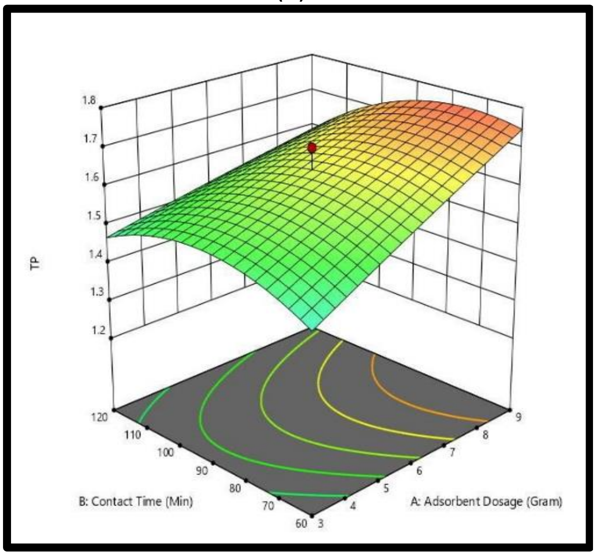
Overall, the concentration of COD is lower at a contact time of 90 minutes than at a contact time of 60 minutes, but it increases at 120 minutes regardless of the mixing speed value. However, the best performance was recorded for 6 g/100 ml CTW at 300 rpm shaking speed and 90 minutes contact time. At this condition, lowest COD concentration was obtained with 80% removal efficiency. CTW showed higher removal of COD as compared steel slag adsorbent that recorded only 55% removal [19]. The highest TP removal (71%) was recorded at 100 rpm shaking speed, 6 g/100 ml CTW dose and 90 minutes contact time. Due to the larger total surface area of the adsorbent and the increasing number of available adsorption sites, the removal concentration increased at higher adsorbent dosages. However as, the shaking speed increased to 200 rpm and 300 rpm, the optimum TP removal was obtained at a lower dosage (3 g/100 ml). Increasing the shaking speed should improve the ability of the contaminants to diffuse into the surface of the adsorbent [10]. However, based on the result of this study the increment of shaking speed changed the optimum dose to 3 g/100 ml however the final concentration of TP was still lower than 9 g/100 ml at100 rpm shaking speed. Thus, it can be concluded that for TP removal, slow shaking speed required higher dose of CTW meanwhile lower dose required increment of shaking intensity as well as reduction of contact time. A study by Yi et al [20] obtained good removal of TP from wastewater as they applied steel slag activated by high temperature. CTW were made from clay that had been through burning process that probably resulted in the removal of TP.



(a)



(b)



(c)

Figure 5. Three-dimension response surface plot for removing (a) pH, (b) COD, (c) TP from LLW using CTW a response if interaction between independent factors.

4. Conclusion

The study performed to optimize removal pH, COD and TP from LLW by using CTW. According to the data analysis, the results discovered that the highest TP removal was 71% with a dosage of 6 g/100 ml ceramic adsorbent, a contact time of 90 minutes, and a shaking speed of 100 rpm. Meanwhile, the highest removal of COD was 80% at a dosage of 6 g/100 mL of ceramic adsorbent, a contact time of 90 minutes, and a shaking speed of 300 rpm. The optimal final concentration value for COD is 60 mg/L and TP is 1.79 mg/L while pH value is 7.13. The major functional group on the CTW were C=C, C=O, C-O=H and were responsible for the removal of the physic-chemical (COD and TP) parameters in LLW. Instead of being disposed of as waste, CTW can be used extensively as adsorbent for various polluted water due to its low cost and good adsorption capacity.

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