INTI JOURNAL | eISSN:2600-7320
Vol.2019:40

Pollutants Removal from Contaminated Water using Agricultural Tea Waste (*Camellia Sinensis*)

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Abstract

The use of natural waste material as an alternative media for treating contaminated river water has gain more popularities in recent years since it is generally low cost and environmentally friendly. This is in line with the 3R campaign promoted by the National Solid Waste Management Department in Malaysia, aiming to overcome the landfill disposal issues. This study presents the investigation of heat-treated tea waste and acid-treated tea waste as an adsorbent to reduce the Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), Total Suspended Solid (TSS), Total Nitrogen (TN) and Total Phosphorus (TP) occurring naturally in contaminated water. The contaminated water sample used in this study was circulated in a prototype to pass through the treated tea waste for five consecutive days. The water parameters were tested in accordance to the APHA method, which were later analysed and compared by evaluating the adsorption capacity, percentage removal of each pollutants and kinetic order reactions. It was identified that two different pre-treatment tea waste follow closely to the pseudo-second-order model due to chemisorption. The results show that the removal of BOD, COD, TP, TN, and TSS by using acid-treated tea waste were 38%, 35%, 68.38%, 42.36%, and 96.31% respectively. Comparatively, heat-treated tea waste was found to be only effective in removing TP, TN, and TSS, with a percentage removal of 53.91%, 78.22%, and 86.84% respectively. Overall, the finding of the study indicates that acid-treated tea waste is potential to be used as a low-cost adsorbent to reduce organic matter, nutrients, and suspended solids in contaminated water.

Keywords

Adsorption, Tea Waste, adsorption kinetic, contaminated water

Introduction

Urbanization and population growth has led to greater demand of freshwater from the Earth water resources. However, these growths have also gradually affect the purity of the water quality due to the increasing amount of waste generated from human activities (Huang et al., 2015). It is generally a challenging task to prevent these hazardous pollutants from entering the water bodies since the pollutants may come from a point-source and non-point source. At presence, a solution for this issue is to acquire a water treatment method in removing the disease-causing agents in the contaminated water (Evans et al., 2012). Among various water treatment methods encountered in the literature search, adsorption was identified to be the best

International Conference on Innovation and Technopreneurship 2019
Submission: 1 June 2019; Acceptance: 19 June 2019

method to treat the pollutants via an effective adsorbent to reduce the pollutants in the contaminated water. It is also regarded as an easier method amongst the various water treatment methods currently available.

The utilization of natural waste materials as optional adsorbent has gained serious interest in removing harmful pollutants from contaminated water resources due to their availability, affordability and eco-friendliness. Statistics have shown that there is approximately 988 million tons of global agricultural waste produced per year and that 1.2 million tons of these agricultural waste are directly disposed into landfills in Malaysia, annually (Emenike et al. 2011). This has caused a very serious impact to the environment, subsequently destroy the fresh water resources due to the leachate of hazardous contaminants like lead, mercury, cadmium and other organic compounds, just to name a few. Therefore, it is encouraged to reduce the amount of solid waste generated especially the agricultural waste in the effort to conserve the landfill disposal area. This indirectly reduces the environmental related issues. An effort demonstrated in this study is to utilize the tea waste as an adsorbent in treating contaminated water. It is selected as the insoluble cell walls are largely made up from cellulose, lignin, tannins and structural protein that make it a potential adsorbent to be used in adsorption method for heavy metal pollutants removal (Singh & Singh, 2012). Nevertheless, it is still lacking of sufficient research works that deal with the removal of organic matter, nitrogen compound, suspended solids and phosphorus compound in contaminated water by using tea waste as the adsorbent. Thus, the present study aims to investigate the effectiveness of heat-treated tea waste and acid-treated tea waste as an adsorbent to reduce organic matter, nutrients, and total suspended solids from contaminated water.

Methodology

Material Preparation

The adsorbent tea waste to be used in this study was collected from various restaurants. Soluble and color components from the raw tea waste were removed by washing repeatedly until they are virtually colorless. The particle size of the tea waste used in the experiment is between 0.35mm to 1.18mm (Zuorro et al., 2013; Pirbazari et al., 2014; Uddin et al., 2009). After washed, the tea waste was dried in the oven at 85 ºC for 12 hours and heat-treated dried tea waste was stored in sealed polyethylene bags (Pirbazari et al., 2014; Girisha, Singha, & Goyala, 2017; Nur-E-Alam, Mia, & Chowdhury, 2017; Zuorro et al., 2013; Uddin et al., 2009). After 12 hours, one batch of tea waste was soaked in 0.1M weak Hydrochloric acid (HCI) for chemical acid treatment for 2 hours and dried in the oven again for another 12 hours at 85 ºC (Tee & Khan, 1988; Girisha, Singha, & Goyala, 2017). The acid chemical pre-treated tea waste was also stored in polyethylene bags and labeled before it is ready to use.

Experimental Works

The water sample was collected from River Batang Labu where the sampling point is located near Kampung Tanjung Nilai, Negeri Sembilan. The prototype, which is equipped with water storage tank, submersible water pump and valves, was used to circulate the water sample by using two different pre-treated tea waste. The water sample was allowed to circulate in the prototype for five continuous days at a constant flow rate. Five different types of water testing parameter were tested, which includes the Biochemical Oxygen Demand (BOD), Chemical Oxygen Demand (COD), Total Suspended Solid (TSS), Total Nitrogen (TN) and Total

Phosphorus (TP). The test were conducted in accordance to American Public Health Association (APHA) method, a standard method for the examination of water and wastewater on a daily basis (APHA, 2005). In addition, a control sample was also prepared for comparison purpose. Similarly, it is allows to circulate in the prototype for five continuous days which later investigated for the different water parameters in the absence of treatment medium. Table 1 shows the standard method used for each water parameters.

Table 1. Standard method for each water parameters according to APHA (APHA, 2005)

Water Parameters	Standard method used
Biochemical Oxygen Demand (BOD)	APHA 5210 B
Chemical Oxygen Demand (COD)	APHA 5220 C
Total Suspended Solid (TSS)	APHA 2540D
Total Nitrogen (TN)	APHA 4500 N_{org} B
Total Phosphorus (TP)	APHA 4500 P B and APHA 4500 P C

Analytics method

Results obtained from the experimental works were analyzed for the adsorbent adsorption efficiency by calculating and evaluating the adsorption capacity and percentage removal for each pollutants. The number of contaminants reduced or adsorbed by the two different types of pretreated tea waste was calculated by using the Equation 1 (Yao & Chen, 2015),

$$A\ (\mathrm{mg/g}) = \frac{(C_i - C_f) \times V}{m} \tag{1}$$

where A is the adsorption capacity expressed in milligram per gram (mg/g), C_i and C_f are the initial and final concentration of pollutants, expressed in milligram per Litre (mg/L), V is the volume of the contaminated water used, expressed in Litre (L), and m is the mass of adsorbent used, expressed in gram (g).

Adsorption kinetic models are used to examine the rate of the adsorption process and potential rate-controlling step. In the present work, the kinetic data obtained from the experimental works were analysed by using pseudo-first order and pseudo-second order models shown in Equation 2 and Equation 3,

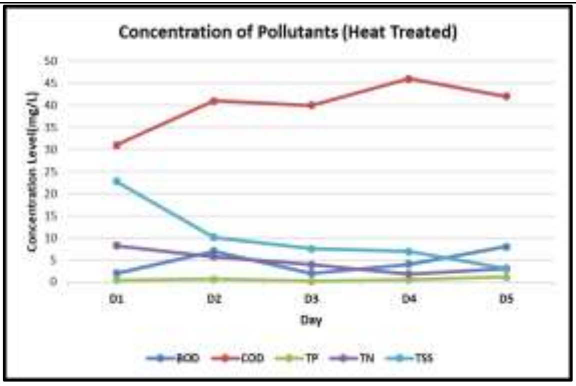
$$\log(q_e - q_t) = \log(q_e) - \frac{k_1}{2.303} t \tag{2}$$

$$\frac{t}{q_t} = \frac{1}{(k_2)(q_e)^2} + \frac{1}{q_e} t \tag{3}$$

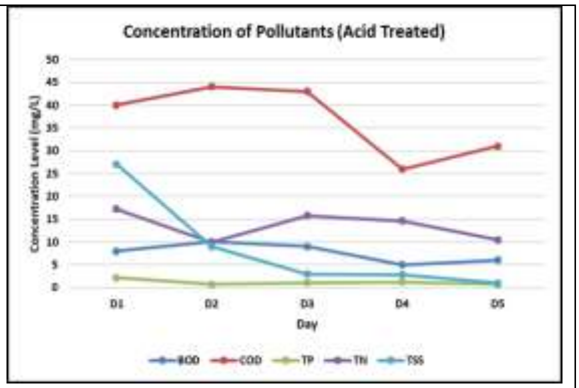
where q_e and q_t are the adsorption density expressed in milligram per gram (mg/g) at equilibrium and at time t, expressed in minutes, respectively. k_1 and k_2 on the other hand, refers to the rate constant of Pseudo-first-order and Pseudo-second-order adsorption.

Results and Discussion

The effect of the contact time, initial concentration of adsorbates and adsorbent dose were studied at the beginning of the experiment. The experimental results on the pollutants removal using heat-treated tea waste and acid-treated tea waste is shown in Figure 1.



(a)



(b)

Figure 1. The concentration of pollutants level from Day 1 to Day 5 using (a) heat-treated tea waste and, (b) acid treated tea waste as adsorbent.

Based on Figure 1 (a), it was observed that the heat-treated tea waste shows only positive pollutants removal for TN, TP, and TSS. The maximum percentage removal of TN, TP, and TSS for the five testing days were 78.22%, 53.91%, and 86.84% respectively. The highest adsorption capacity recorded for TN, TP, and TSS were 4.293mg/g at Day 4, 0.138mg/g at Day 3, and 13.2mg/g at Day 5. As in Figure 1 (b), the maximum percentage removal of pollutants using acid-treated tea waste for the five testing days on BOD, COD, TP, TN, and TSS were 38%, 35%, 68.38%, 42.36%, and 96.31% respectively. The adsorption capacity of acid-treated tea waste returns highest adsorption capacity of 2mg/g and 9.333mg/g for BOD and COD at Day 4, 0.983mg/g and 4.864mg/g for TP and TN at Day 2 and 17.4mg/g at Day 5 for TSS adsorption.

The fluctuation of the organic matter and nutrients removal using tea waste could be related to the presence of microorganisms in the naturally contaminated water (Cai et al., 2019). The acid-treated tea waste possessed an ability to purify the tea waste, resulting in an increment of total acid content in the tea waste. This in return results in an increase of percentage removal in BOD. Based on the previous study conducted by Powell (2003), the acid content in the tea waste is very proficient in removing microorganisms hence reducing the microorganism decomposition and increase the percentage of BOD removal. On the other hand, the contaminated water that was circulating through the tea waste may caused various sizes of particles, algae, microorganisms and other inorganic matter to be trapped in the pores of the tea waste that slowly formed a biofilm layer on the surface of the adsorbent to further reduce the concentration level of COD (Zhang et al.,2018). Adsorption of TP is also significantly dependent on the pH value of the water; the extent of phosphorus adsorbed in unit adsorbent would increase with the lower pH value (Tu et al., 2016).

The heat-treated and acid-treated tea waste also shows effective removal of TN. It was identified that the heat-treated tea waste seems to be more efficient in TN adsorption as compared to the acid-treated tea waste. The TN defined in this study represents the sum of organic and inorganic nitrogen in raw water, which includes nitrites, nitrates, and ammonia.

Zuorro et al., (2013) identified that nitrification and denitrification processes can help to reduce the concentration level of TN, which is actually happening in this experiment. These processes are further strengthen in the presence of structural proteins of tea waste like the hydroxyl groups and carboxyl groups on the cellulose fibers apart from the availability of phenolic compounds in the proteins.

In TSS removal, heat-treated tea waste and acid-treated tea waste is capable of removing 86.84% and 96.31% of suspended solids at Day 5. This is associated to the micro pores presence within the tea waste, which have the ability to trap the suspended solids and dissolved solids from the contaminated water circulating through the adsorbents (De Gisi et al., 2016). It was also identified from the previous study that acid-treated tea waste can increased the porosity of tea waste, which explains the fact that the percentage removal of TSS is higher compared to heat-treated tea waste (Pal et al., 2019).

The adsorption characteristics were also studied in the present work. Table 2 shows the finding by using Pseudo-first-order model and Pseudo-second-order model for heat-treated tea waste and acid-treated tea waste.

Table 2. Pseudo-first-order and Pseudo-second-order model for heat-treated tea waste and acid treated tea waste.

Heat-treated Tea Waste						**Acid-treated Tea Waste**					
	BOD	**COD**	**TP**	**TN**	**TSS**		**BOD**	**COD**	**TP**	**TN**	**TSS**
Pseudo-first-order											
R^2	0.1917	0.3033	0.1051	0.1935	0.0186	**R^2**	0.209	0.001	0.7975	0.0191	0.8027
K_1	-0.0078	-0.0099	0.006	0.0051	0.0025	**K_1**	0.0083	0.0002	0.0322	0.0035	0.0062
q_e	1.2266	0.8831	0.7096	1.6244	2.3201	**q_e**	9.049	27.657	1.245	1.05	1.032
Pseudo-second-order											
R^2	0.702	0.917	0.161	0.853	0.955	**R^2**	0.820	0.693	0.892	0.604	0.987
K_2	0.081	-0.036	0.611	0.018	0.01	**K_2**	-0.019	-0.004	0.455	0.328	0.056
q_e	-2.532	-8.065	-0.165	4.098	12.97	**q_e**	1.212	4.926	0.821	3.049	17.857
Experimental qe (mg/g)											
	0	**-6**	**-0.49**	**4.29**	**13.2**		**2**	**9.33**	**0.98**	**4.86**	**17.40**

Based on Table 2, Pseudo-second-order equation better fits the experimental data with correlation coefficient (R^2) closer to 1. The experimental maximum adsorption capacity (q_e) is also identical to the calculated maximum adsorption capacity (q_e) upon comparing with the pseudo-first-order model for all the water parameters. Such finding indicates an inclination towards chemisorption where there existed a chemical bond on the surface of the adsorbent to bind the adsorbate (Kandisa & Saibaba, 2018). The fact that the adsorption kinetics better fits the pseudo-second-order model is expected since the initial concentration of contaminated water is low (Moussout et al., 2018). The values of K_2 in the experiments represent the operating conditions such as initial pH and solution concentration, temperature, and agitation rate. It decreases as the initial solution concentration increases since it takes a longer time to reach to an equilibrium.

Conclusions

The present study aim to reduce organic matter, nitrogen compound, suspended solid and phosphorus compound from contaminated water by using agricultural tea waste which were pre-treated with heat and acid. The result shows when acid-treated tea waste was used as the adsorbent, the percentage removal of BOD, COD, TP, TN, and TSS are 38%, 35%, 68.38%, 42.36%, and 96.31% respectively. Comparatively, heat-treated tea waste only managed to remove 53.91%, 78.22%, and 86.84% of TP, TN and TSS during the five days adsorption experiment. The Pseudo-second-order equation fitted the experimental data well for both heat-treated tea waste and acid-treated tea waste. This indicates that both reactions are more inclined towards chemisorption characteristics. Within the scope of the study, it is concluded that acid-treated tea waste is more efficient in pollutants removal as compared to heat-treated tea waste.

Acknowledgment

The authors are grateful to INTI International University for the Final Year Project financial support in this project.

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