International Journal of Engineering & Technology, 7 (4.18) (2018) 256-261

International Journal of Engineering & Technology

Website: www.sciencepubco.com/index.php/IJET

Research paper

Biosorption of coloured wastewater onto citric acid modified *casuarina equisetifolia* leaves powder

Siti Fatma Binti Abd Karim [1]*, Nurul Syahirah Binti Abd Kahar [1], Ummi Kalthum Ibrahim [1], Rabiatul Adawiyah Abdol Aziz [1]

[1]*Fakulti Kejuruteraan Kimia, Universiti Teknologi Mara, 40450 Shah Alam, Selnagor, Malaysia*
**Corresponding author E-mail: sitifatma9633@salam.uitm.edu.my*

Abstract

Biosorption is a process that utilizes dead or living biomass and particularly useful for the removal of industrial contaminants. Pine needle (*Casuarina Equisetifolia*) leaves are an agriculture waste material and can be found abundantly in Malaysia and all over the world. The objectives of this research are to prepare the untreated and treated biosorbent from *Casuarina Equisetifolia* leaves and to determine the effect of pH, dosage of biosorbent, contact time and initial concentration of the solution to the colour removal. The feasibility potential of treated *Casuarina Equisetifolia* leaves powder is studied by using a batch mode method that investigates the pH, dosage of biosorbent, contact time and initial concentration of the solution to the colour removal. Samples were analyzed using UV-Visible Spectrophotometer (UV-Vis), Fourier Transform Infra-Red Spectroscopy (FTIR) and Scanning Electron Microscope (SEM) to determine the biosorption capacity. It was observed that treated *Casuarina Equisetifolia* leaves powder able to achieve 99.72% of colour removal efficiency at optimum condition where pH is 6, dosage of biosorbent is 1.0 g, contact time is 60 minutes and initial concentration of the solution is 10 ppm. This research also observed that treated *Casuarina Equisetifolia* leaves powder results in an attractive alternative for removing colour that shows a great reduction of coloured wastewater.

Keywords: *Biosorbent; Biosorption; Casuarina Equisetifolia (CE) leaves; Methylene blue (MB)*

1. Introduction

Coloured wastewater is a result of batch processes both in the dye manufacturing industries and in the dye-consuming industries. The textile industry is one of the most complicated industries among manufacturing industry. It produces a large amount of dye effluents, which are highly toxic as they contain a large number of metal complex dyes. Strong colour of the textile wastewater is the most serious problem of the textile industrial effluents. Many operations of the textile process are water intensive in textile production. Therefore, a purification treatment to recycle water must have better performance than for simple discharge according to the limits imposed by legislation. There are a group of processes available for the removal of dyes by conventional treatment technologies including biological and chemical oxidation, coagulation and biosorption but they cannot be effectively used individually. Powdered activated carbon is used mainly for water treatment, production of industrial chemicals and beverage clarification. However, some textile industries do not treat their wastewater by using activated carbon because of the high market price that varies directly as it is a function of demand, quality of the product and cost of production [1].
The colour removal technologies can be classified into three categories which are biological, chemical and physical [2]. Biological treatment process is often proved to be the most economical as compared to physical and chemical treatment processes. Bhattacharyya and Sharma found that the biological treatment has flexibility in design and operations [3]. However, it is constrained towards the toxicity and diurnal variation and also requires a large land area. Chemical methods are accommodating in the removal of dyes from the wastewater. These techniques are costly and even though the dyes are removed, the addition of concentrated sludge creates a disposal problem [4]. There is also the opportunity that a secondary pollution problem will occur because of too much chemical use.
Physical methods such as membrane filtration and biosorption are widely used. The major disadvantage of membrane filtration processes is that they have an inadequate lifetime and the cost of periodic replacement must also be integrated in any analysis of their economic capability. It was reported that biosorption process is an effective and best equilibrium process for the removal of decontaminants from the wastewater [5]. This process provides an option for the treatment of contaminated waters especially if the sorbent is economical and does not involve any additional pre-treatment step before its application. An important number of biosorption studies have been focussed to optimizing biosorption parameters in order to obtain the maximum removal efficiency.
Biosorption is measured as a fast physical or chemical process [6]. An important number of biosorption studies have been focused on optimizing biosorption parameters in order to obtain the maximum removal efficiency. Biosorption process is a very complicated process. The binding of metal ions by natural materials may possibly take place through two types of biosorption process which are physical (electrostatic interaction and van der Waals forces) or chemical (ion exchange) process. According to the researchers, the composition of the cell wall is of great significance to the biosorption process [7].

The cost of treatment will decrease if the biosorbents is made up from inexpensive alternative biosorbents such as orange peel, banana peel, lemon peel, peanut hull and coir pith [8]. Table 1 shows the comparison of biosorption capacity of several low cost biosorbents for the uptake of methylene blue dye from its aqueous solutions.

Table 1: Comparison of biosorption capacity of several low cost biosorbents for the uptake of methylene blue dye from its aqueous solutions [8]

No	Type of Biosorbents	Biosorption Capacity (mg/g)	References
1.	Rice husk	40.59	(Vadivelan & Kumar 2005)
2.	Neem leaf powder	8.76	(Bhattacharyya & Sharma 2005)
3.	Banana peel	20.8	(Annadurai et al. 2002)
4.	Orange peel	18.6	(Annadurai et al. 2002)
5.	Raw date pits	80.3	(Banat et al. 2003)
6.	Peanut hull	68.03	(Gong et al. 2005)
7.	Coir pith	120.43	(Namasivayam et al. 2001)
8.	Fly ash	5.57	(Kumar et al. 2005)
9.	Glass fibers	2.24	(Charabarti & Dutta 2005)
10.	Clay	6.3	(Gurses et al. 2004)
11.	Diatomite	156.6	(Al-Ghouti et al. 2003)
12.	Activated sludge	256.41	(Gulnaz et al. 2004)
13.	Aspergillus niger	18.54	(Fu & Viraraghavan 2000)
14.	Cotton waste	277	(McKay et al. 1999)
15.	Neem sawdust	2.1-3.6	(Khattri & Singh 2000)
16.	Lemon peel	29	(Kumar & Porkodi 2006)
17.	Palm kernel fiber	217.96	(Ofomaja, 2007)
18.	Jackfruit	252.83	(Tamez et al. 2009)
19.	Ginkgo leaf	39	(Mu et al. 2011)
20.	Silica	11.21	(Woolard et al. 2002)
21.	Defatted papaya seed	1250	(Unuabonah et al. 2009)
22.	Undefatted papaya seed	769.23	(Unuabonah et al. 2009)
23.	Boiled papaya seed	555.55	(Hameed, 2009)
24.	Papaya leaf powder	512.55	(Zobayer et al. 2012)

Therefore, this research is conducted to determine the efficiency of biosorption of coloured wastewater onto citric acid modified *Casuarina Equisetifolia* leaves powder. In this research, *Casuarina Equisetifolia* leaves used is collected at the beaches or near the coast on sandy soils and methylene blue dye solution is selected as a sample of coloured wastewater. This research will focus on determining the optimum pH condition (2, 3, 4, 5, 6, 7), dosage of biosorbent (0.1 g, 0.3 g, 0.5 g, 1.0 g, 1.1 g, 1.3 g), contact time (15 minutes, 30 minutes, 60 minutes, 90 minutes) and initial concentration of the solution (10 ppm, 50 ppm, 100 ppm, 150 ppm). This condition will determine the percent for colour removal that absorb by *Casuarina Equisetifolia* leaves.

2. Methodology

2.1. Materials

All reagents were of analytical grade chemicals. All solutions were prepared with distilled water. All of the chemicals used in this study were purchased from R & M Chemicals including the cationic dye, MB and the modified agent, citric acid.

2.2. Biosorbent

CE leave powder was used as biosorbent to remove MB dye from aqueous solution. CE leaves were collected at Pantai Cahaya Bulan, Kota Bharu, Kelantan. Upon collection, the leaves were washed and rinsed with distilled water for few times to remove dust and impurities. The leaves were boiled with distilled water at 100 °C for 3 hours until it is free from colour and turbidity. Then, the leaves were air dried and subsequently oven dried in the universal oven (Model: UFE600 MEMMERT) at 100 °C for 24 hours to remove any moisture from the leaves. Prior to use, the dried leaves were powdered using a grinder machine (Model: DICKSON DFY-200). The leaves were passed through digital sieve shaker (Model: OCTAGON 2000) with diameter 125 µm used for biosorption studies. The untreated leaves powders were kept in airtight plastic and stored in the desiccators (Model: DURAN).
The CE leaves powders were treated with citric acid. It is prepared by soaking 30 g of readily CE leaves powder into citric acid solution and stirred for 24 hours. It is then air dried and subsequently oven dried at 50 °C for 24 hours. The dried treated CE leaves powders were kept in airtight plastic for further use.

2.3. Chemical preparation

MB dye solution was selected as the sample of coloured wastewater. A stock solution of the MB dye is prepared by dissolving accurately weighted amount of MB powder in distilled water. Appropriate dilution of the stock solution is carried out to obtain desired concentration of MB dye solution.

2.4. Biosorption studies

Four parameters were determined in this study for untreated and treated CE leaves powders which are pH, dosage of biosorbents, contact time of biosorbents with the MB dye solution and initial concentration of MB solution. All of four parameters are related to each other. The ultraviolet visible (UV-Vis) (Model: Perkin Elmer LAMBDA 750) used to determine the wavelength and maximum absorbance of biosorbent. The percentage of colour removal for each of parameters studied was calculated using the equation below:

$$\text{Percent of colour removal} = [(C_{inital} - C_{exit}) / (C_{initial})] \times 100\% \text{ acid.} \quad (1)$$

2.5. Fourier transform infrared (FTIR) spectroscopy

Fourier transform infra-red spectroscopy (FTIR) of the biosorbent was obtained (and transform to Microsoft Excel) by using an FTIR spectrophotometer (Model: Perkin Elmer Spectrum One) covering a wave number range of 400-4000 cm^{-1} was used to identify the chemical components of the leaves.

2.6. Scanning electron microscopy (SEM)

Scanning electron microscope (SEM) (Model: QUANTA FEG 450) was used to characterized the morphology surface of biosorbent. The SEM micrograph used was at 5000 x magnification.

2.7. Experimental set-up for biosorption process of colour removal

To characterize the CE leaves, a number of basic properties were determined. Since coloured wastewater is very sensitive to pH, parameter for pH experiment was started first. The pH of each sample was measured using a desktop pH meter (Model: Mettler Toledo). Six different beakers of 50 ml of MB dye solution were fixed to 50 ppm of concentration. Then, the pH for six different beakers were adjusted into six different pH values which are pH 2.0, 3.0, 4.0, 5.0, 6.0 and 7.0 for each of the beaker. Next, 0.3 g of CE leaves powder was added into each of the solutions that contain six different pH values. Each of the mixture was shaken at 30 °C for 60 minutes at the speed of 100 rpm in the incubator shaker (Model: INFORS HT ECOTRON) simultaneously. Then, the CE leaves powder was centrifuged for 15 minutes at speed of 5000 rpm in order to ensure that it is sedimented at the bottom of each tube using a benchtop refrigerated centrifuge (Model: SIGMA 3-18K). Next, the colour intensity of MB dye solution was determined in the filtrates by using UV-Vis. After that, the percent for colour removal for each sample was calculated and recorded.

For the second parameter which is dosage of biosorbent, six different beakers of 50 ml of MB dye solution were fixed to the 50 ppm of concentration with pH value that has the highest percent removal obtained from the previous parameter. Then, six different mass of CE leaves powder which are 0.1 g, 0.3 g, 0.5 g, 1.0 g, 1.1 g and 1.3 g were added into each of six different beakers. Each of the mixture was shaken at 30 °C for 60 minutes at the speed of 100 rpm in the incubator shaker simultaneously. Then, the CE leaves powder was centrifuged for 15 minutes at the speed of 5000 rpm in order to make sure that it is sedimented at the bottom of each tube using the benchtop refrigerated centrifuge. Next, the colour intensity of MB dye solution was determined in the filtrates by using UV-Vis. After that, the percent for colour removal for each sample was calculated and recorded.

For the third parameter which is contact time, four different beakers of 50 ml of MB dye solution were fixed to the 50 ppm of concentration with pH value that has the highest percent removal obtained from the first parameter. Then, the dosage of biosorbent that has the highest percent removal from the second parameter was added into each of the four beakers. Each of the mixtures was shaken at 30 °C at the speed of 100 rpm in the incubator shaker simultaneously with four different contact times which are 15 minutes, 30 minutes, 60 minutes and 90 minutes. Then, the CE leaves powder was centrifuged for 15 minutes at the speed of 5000 rpm in order to make sure that it is sedimented at the bottom of each tube using the benchtop refrigerated centrifuge. Next, the colour intensity of MB dye solution was determined in the filtrates by using UV-Vis. After that, the percent for colour removal for each sample was calculated and recorded.

For the last parameter which is the initial concentration, four different beakers of 50 ml of MB dye solution were fixed with four different concentrations of the solution which are 10 ppm, 50 ppm, 100 ppm, and 150 ppm with pH value that has the highest percent removal obtained from the first parameter. Then, the dosage of biosorbent that has the highest percent removal from the second parameter was added into each of the four beakers. Each of the mixtures was shaken at 30 °C for 60 minutes at the speed of 100 rpm in the incubator shaker simultaneously. Then, the CE leaves powder was centrifuged for 15 minutes at the speed of 5000 rpm in order to make sure that it is sedimented at the bottom of each tube using the benchtop refrigerated centrifuge. Next, the colour intensity of MB dye solution was determined in the filtrates by using UV-Vis. After that, the percent for colour removal for each sample was calculated and recorded.

All of the four parameters were tested using both untreated and treated CE leaves powder and the highest percentage of colour removal is recorded.

3. Results and discussion

3.1. Fourier transform infrared (FTIR) spectroscopy

The FTIR spectra for untreated CE leaves powder is shown in Fig. 1 and Table 2 while for treated CE leaves powder is shown in Fig. 2 and Table 3.

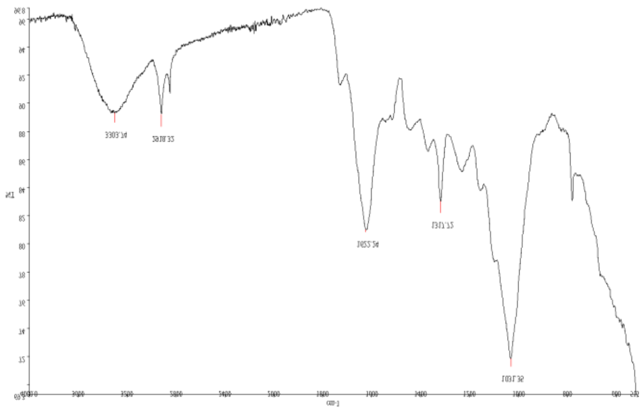

Fig. 1: FTIR spectra of untreated CE leaves powder in the region from 400 cm^{-1} to 4000 cm^{-1}.

Table 2: FTIR spectra data of untreated CE leaves powder

No	Adsorption Peak (cm^{-1})	Bond	Type of compound
1	3303.74	O-H	Phenol
2	2918.32	O-H	Carboxylic Acid
3	1622.24	C=C	Aromatic

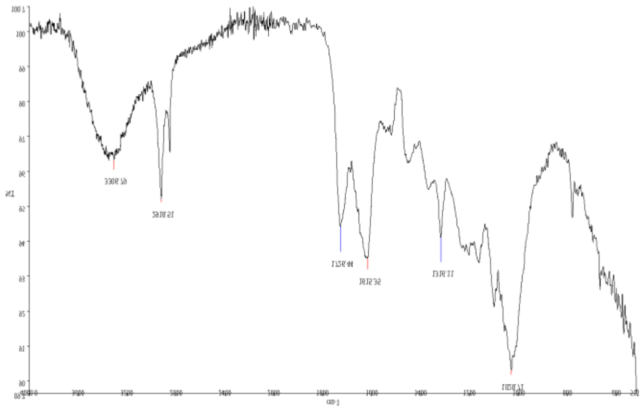

Fig. 2: FTIR spectra of treated CE leaves powder in the region from 400 cm^{-1} to 4000 cm^{-1}.

Table 3: FTIR spectra data of treated CE leaves powder

No	Adsorption Peak (cm^{-1})	Bond	Type of compound
1	3306.79	O-H	Phenol
2	2918.51	O-H	Carboxylic Acid
3	1726.44	C=O	Carboxylic Acid
4	1615.35	C=C	Aromatic

According to Fig. 1 and Fig. 2, both untreated and treated CE leaves powder showed the presence of –OH bond which meant they are negatively charge. Since MB dye is cationic dye, it is possible for CE leaves powder to absorb the colour from the MB dye solution. However, the treated CE shows additional presence of C=O bond located at 1726.44 cm^{-1} was characteristics of carboxylic acid stretching. It is understood that this presence is the reason that the treated CE is treated with citric acid solution and the functional group for citric acid is carboxylic acid with condensed formula -COOH. In addition, the presence of C=O bond has made the treated CE stronger and contain more negative charge which is able to absorb more colour from the MB dye solution compared to untreated CE.

3.2. Scanning electron microscopy (SEM)

The SEM micrograph for untreated CE leaves powder is shown in Fig. 3 and Fig. 4 while for treated CE leaves powder is shown in Fig. 5 and Fig. 6.

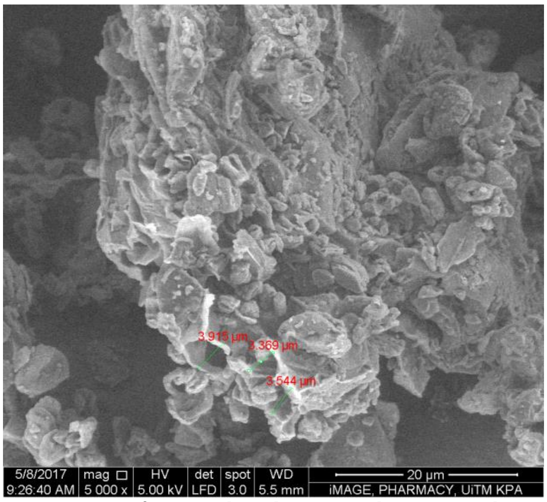

Fig. 3: SEM image of untreated CE leaves powder micrograph at 5000 x magnification.

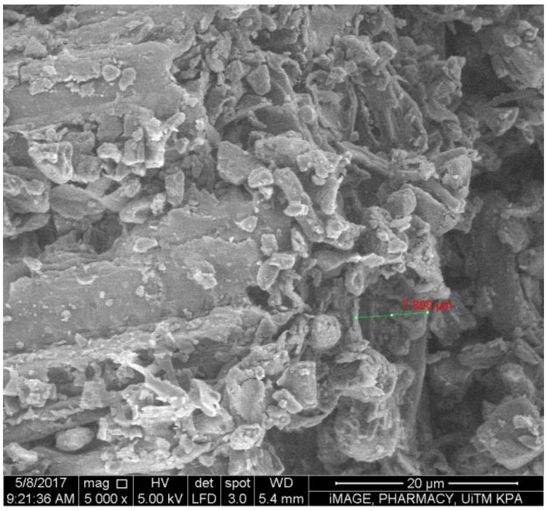

Fig. 4: SEM image micrograph at 5000 x magnification

Fig. 3 shows several length of pore size presented which are 3.369 µm, 3.544 µm and 3.915 µm. The longest length of pore size presented in untreated CE leaves powder which is 7.890 µm was presented in Fig. 4 while Fig. 5 shows several small pores of different length appeared in which are 1.877 µm, 2.346 µm, 2.836 µm, 4.418 µm, 5.565 µm, 6.353 µm, 7.397 µm and 7.882 µm. The longest length of pore size presented in treated CE leaves powder which is 17.46 µm as shown in Fig. 6. Based on the SEM image in Fig. 3, Fig. 4, Fig. 5 and Fig. 6, it clearly shows that treated biosorbent depicted more porosity compared to untreated biosorbent. The small pores of different length obtained in the treated CE provide suitable binding sites for MB solution. The higher the number of biosorbent pore, the higher the rate of absorption into the biosorbent [9]. The higher number of pores that existed in the treated biosorbent has proven that treated biosorbent will able to absorb more MB dye solution compared to untreated biosorbent. The different length of small pores and the higher number of pores presented in the treated biosorbent proved that treated biosorbent is more efficient to be used in the colour removal application in the industry.

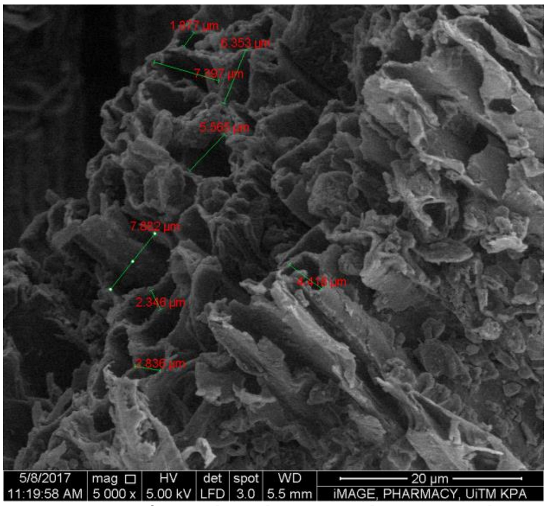

Fig. 5: SEM image of treated CE leaves powder micrograph at 5000 x magnification

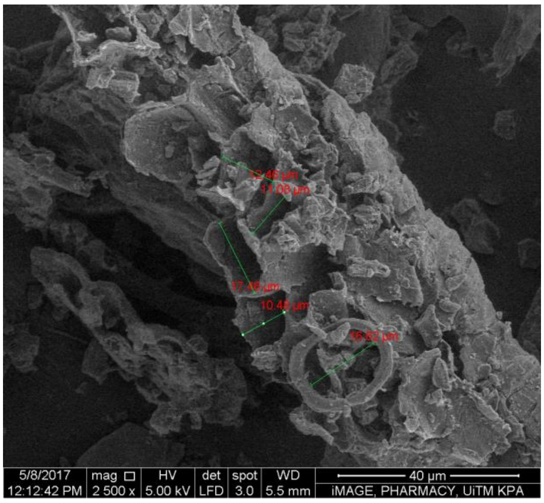

Fig. 6: SEM image micrograph at 5000 x magnification.

3.3. Effect of pH

In biosorption process, pH plays an important role in determining the biosorbent efficiency. Each biosorbent is very sensitive over the range of pH. So, the suitable pH needs to be identified in order to get high biosorbent efficiency.

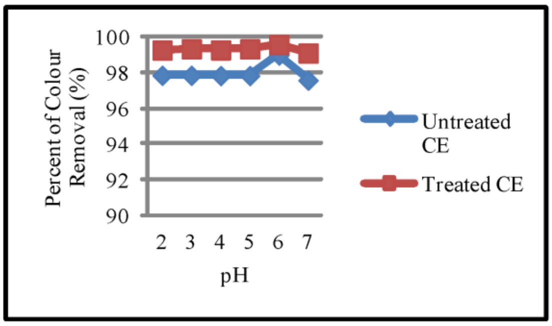

Fig. 7: Graph of percent removal for both untreated and treated CE leaves at different values of pH.

Based on the graph shown in Fig. 7, it is proven that treated CE has obtained higher percent of colour removal compared to untreated CE. From this research, it is also proven that the higher the value of pH of the dye solution, the higher the percent of colour removal of the dye solution. However, the percent decreased once the pH of the dye solution reached pH 7 because the optimum condition is already reached. Thus, this research shows that the acidic medium is favourable for CE leaves powder

for biosorption process of MB dye solution and is not suitable to use at neutral condition since it will produce less percent removal.
It was found that for pH less than 6, a significantly high electrostatic force of attraction exists between the negatively charged surfaces, hence enhancing the dye uptake [10]. However, with the increase of pH, the negatively charged sites of the biosorbent get decreased and the surface of the biosorbent became positively charged. This condition does not favour the uptake of anionic dye from the system and leads to electrostatic repulsion.

3.4. Effect of biosorbent dosage

Biosorbent dosage also plays an important role in this research in order to determine the percent of colour removal of MB dye solution. The effect of biosorbent dosage was investigated by using pH value that has the highest percent removal obtained from the previous parameter.

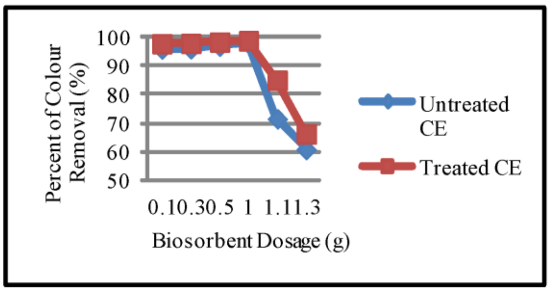

Fig. 8: Graph of percent removal for both untreated and treated CE leaves at different values of biosorbent dosage.

Based on the graph plotted in Fig. 8, it is revealed that treated CE has achieved the greatest percent of colour removal compared to untreated CE. From this research, it is proven that the larger the amount of biosorbent dosage applied to the MB dye solution, the higher the percent of colour removal is achieved. The percent of colour removal decreased at 1.1g of biosorbent dosage was added and keep decreasing at 1.3g of biosorbent dosage. Thus, this research also proved that CE leaves powder is not suitable to be used in the amount of more than 1.0 g because the optimum condition is already reached.
The higher amount of biosorbent dosage will results in a higher percentage of colour removal [11]. The positive correspondence between the adsorbent dosage and dye removal percentage could be attributed to the increase in adsorption sites as well as an increase in the surface area of the biosorbent. However, further increment in the dosage did not give any significant changes in the percentage removal and this could be due to the saturation of the binding sites.

3.5. Effect of contact time

In biosorption process, contact time plays an important role in finding the percent of colour removal of the MB dye solution. The effect of contact time was investigated by using pH value and biosorbent dosage that has the highest percent removal obtained from previous parameters.

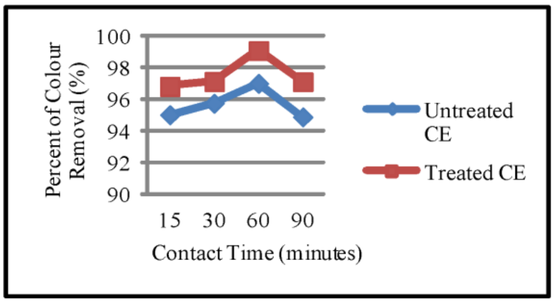

Fig. 9: Graph percent removal for both untreated and treated CE leaves at different contact time.

Based on the graph shown in Fig. 9, it is proven that treated CE obtained a higher percentage of colour removal compared to untreated CE. From this research, it is also established that the higher the contact times of biosorbent with MB dye solution, the higher the percent of colour removal but the percent of colour removal will decrease when the contact time is increase more than 60 minutes because the equilibrium condition is already reached.
Dye exhibited a fast biosorption rate during the first 60 minutes of contact time due to a great accessibility of binding sites for dye molecules to be biosorbed [11]. Usually, the dye anions will attach to all of the active sites until they are fully engaged. Hence, with time, smaller quantities of active sites are accessible and thus decrease the amount of dye being adsorbed.

3.6. Effect of initial concentration

The initial MB dye concentration plays an important role in affecting the capacity of MB dye to absorb onto biosorbents. The effect of initial concentration was investigated by using pH value, biosorbent dosage and contact time that has the highest percent removal obtained from previous parameters.

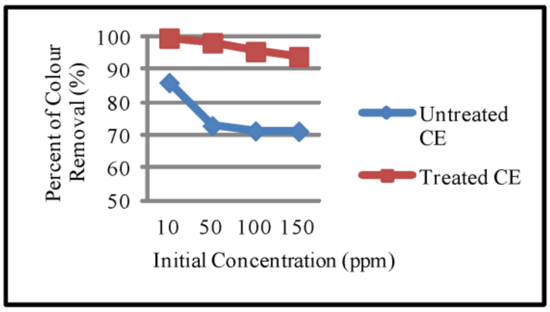

Fig. 10: Graph of percent removal for both untreated and treated CE leaves at different values of initial concentration.

Based on the graph shown in Fig. 10, it is revealed that treated CE achieved the greatest percentage of colour removal compared to untreated CE. From this research, it is shown that the lower the initial concentration of MB solution, the higher the percent of colour removal. It is understood that it is hard to remove colour from high concentration of dye solution because the colour of the solution is darker compared to lower concentration of dye solution which the colour of the solution is much lighter.
The higher the methylene blue concentration, the stronger the driving forces of the concentration gradient and therefore the biosorption capacity is higher [12]. However, higher initial methylene blue concentration will result in lower diffusion efficiency and more competition of biosorption ions for a fixed activated surface site.

4. Conclusion

As a conclusion, from all of these four parameters studied, it is clearly shown that treated CE obtained a higher percentage of colour removal compared to untreated CE. Also, the analysis of FTIR and SEM for both of biosorbents also proved that treated CE has higher potential to absorb colour compared to untreated CE due to the existence of some functional groups and the different length of small pores also the higher number of pores in the treated CE. Thus, after several aspects that have been taken which are pH, biosorbent dosage, contact time, initial concentration of dye solution, the optimum condition of both untreated and treated CE can be determined. The optimum condition for both untreated CE and treated CE is at pH 6, 1.0 g of biosorbent dosage, 60 minutes of contact time between biosorbent and dye solution, 10 ppm initial concentration of dye solution. Therefore, it is shown that both of the biosorbent can be used as a medium in colour wastewater removal but the treated biosorbent is the procedure of choice.

From this research, there are several recommendations that can be used in the future research which includes using other raw material as biosorbent such as rice husk, banana peel, palm kernel fiber and other agricultural waste in order to compare the percent of colour removal obtained from each of them. Also, using other modified agent such as acid and base to treat the raw material use are also recommended for future study. Other than that, other important analysis such as Field Emission Scanning Electron Microscope (FESEM) also can be done for the next future study in order to study the characteristics of the biosorbent used.

Acknowledgement

Special appreciation to Ministry Education of Malaysia and Univeristi Teknologi Mara, for the research grant 600-IRMI/MYRA 5/3/LESTARI (0145/2016) and Fakulti Kejuruteraan Kimia, UiTM Shah Alam for supporting the research project during this study work.

References

[1] Gupta V.K., Mittal A. and Gajbe V. (2005). Adsorption and desorption studies of a water soluble dye, Quilnoline Yellow, using waste materials. Journal of Colloid and Interface Science 284(3): 89-98.

[2] T.Robinson, G. McMullan, R. Marchant, P. Nigam. (2001). Remediation of dyes in textiles effluent: a critical review on current treatment technologies with a proposed alternative. Bioresource Technology 77(1): 247-255.

[3] K.G. Bhattacharyya and A. Sharma. (2003). Adsorption characteristics of the dye, Brilliant Green. Chemical Engineering Journal 84(5): 286-291.

[4] Crini, G. (2005). Recent developments in polysaccharide-based materials used as adsorbents in wastewater treatment. Progress in Polymer Science 30(1): 38-70.

[5] A. Dabrowski. (2001). Adsorption: from theory to practice. Advances in Colloid and Interface Science 93(7): 135-224.

[6] G.M. Gadd. (1990). Heavy metal accumulation by bacteria and other microorganisms Experentia. Chemical Engineering Journal 46(2): 834-840.

[7] M. Izabela, C. Katarzyna and W, K. Anna (2013). State of the art for the biosorption process. Applied Biochemistry and Biotechnology 170(6): 1389–1416.

[8] Md. Tamez Uddin, Md. Rukanuzzaman, Md. Maksudur Rahman Khan and Md. Akhtarul Islam. (2009). Adsorption of methylene blue from aqueous solution by Jackruit (Artocarpus heteropyllus) leaf powder: A fixed-bed column study. J Environ Manage 90(11): 3443-3450.

[9] Lawrence M.A and David R.C. (2015). Reviews in mineralogy and geochemistry.

[10] A.S. Ozcan. (2004). Adsorption of acid dyes from aqueous solutions onto acid activated bentonite. J. Colloid Interf. Sci. 276(2): 39-46.

[11] Zahra Haddadian, Mohammad Amin Shavandi, Zurina Zainal Abidin, Ahmadun Fakhru'l-Razi And Mohd Halim Shah Ismail (2012). Removal Methyl Orange from Aqueous Solutions Using Dragon Fruit (*Hylocereusundatus*) Foliage. Chemical Science Transactions.

[12] C.H. Weng, Kadirvelu K and Panda GC. (2009). Journal of Hazardous Materials 170(23): 417–424.