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Feasibility of Nile tilapia (*Oreochromis niloticus*) *fish scales as biosorbents for dye removal*

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ABSTRACT
Many dyes are used in many industries such as food, paper, carpets, rubbers, plastics, cosmetics and textiles in order to color their product. Dyes are difficult to degrade and could cause potential hazards to the water body and environment due to its complex structures. Biosorption has been found as an effective and eco-friendly technique.The potential of using *Oreochromis niloticus* (Nile Tilapia) fish scale as biosorbent of methylene blue dye removal was investigated. The scales were washed, sun dried, pulverized and sieved before exposed to the simulated wastewater. The effect of operational parameters such as contact time (180 minutes), temperature (30 and 50C), and biosorbent dosages (0.1, 0.2 and 0.3 g/100 ml) was studied in two different simulated wastewater which are from distilled water and industrial wastewater. The extent of dye removal decreased with increasing adsorbent dosage, increased with increasing contact time for both simulated wastewater and increased with decreasing temperature. The biosorption data followed Langmuir isotherm model. The highest removal was achieved at 30C and 0.2g of biosorbent dosage. The results suggest that an eco-friendly, cost-efficient *O. niloticus* fish scales could be used as an effective biosorbent for the removal of methylene blue dye from simulated wastewater.

Keywords: Biosorption, Fish Scales, Methylene Blue Dye, Biosorption Capacity, Langmuir Isotherm

1. Introduction

Dyes are complex chemical compounds used in many industries such as food, paper, carpets, rubbers, plastics, cosmetics, and textiles in order to color their product. Among the many significant uses, it is also a source of challenge in the environment. Dyes are considered important pollutants and chemicals that are resistant to weather, action of detergents, and non-biodegradable. The discharge of untreated colored wastewater from those industries into natural streams is a critical worldwide environmental concern. It becomes one of the main sources of severe water pollution and cause of many significant problems (Saxena et al., 2017).

The presence of dyes in effluents may still remain even after extensive treatment. It causes damage to the ecosystem and also affects the groundwater system due to leaching from the soil (Vital et al.). In a water body, it extremely decreases oxygen concentrations due to the presence of sulfuric acid groups in their molecules that block the passage of light that will not just harm water, but the ecosystem as a whole, making people and aquatic life prone to chemical exposure. In addition, dyes and their metabolites are toxic, mutagenic and carcinogenic and pose potential health hazards to all forms of life. They are resistant to aerobic digestion and signify one of the difficult groups to be removed from the industrial wastewater. That is why wastewater containing dyes are very difficult to treat and a major challenge (Vieira et al., 2011).

Numerous methods are evidently relevant to the dye removal of industrial wastewater. Different methods such as physicochemical processes, biological processes, nuclear treatments, and acoustical, electrical, and electromagnetic processes are available (Gupta et al., 2009; Rafatullah, 2010). Among them the sorption process from physicochemical processes is now considered as the most effective and economical way to treat wastewaters with high content of color (Uzunoğlu et al., 2016). Adsorption technology is especially a favorable option and can prove to be attractive. A large variety of non-conventional adsorbent materials has been proposed and studied for their ability to remove dyes. Activated carbon is well known as the most widely used adsorbent for removal of dye but because of its high cost, this limits its usage in large scale production (X.S. Wang et al., 2008). Meanwhile, a new, cost effective and more environmentally friendly method, the biosorption process is proven to be a favorable process to remove dyes and highly efficient technology for wastewater remediation (Gupta et al., 2008; Fu et al., 2000).

In the Philippines, *Oreochromis niloticus* (Nile Tilapia) is the most cultured freshwater fish and the second most important fish species that is farmed to improve food security and alleviate poverty. Fish scales are very common residues that are being generated from fish markets everyday and are released into the environment causing pollution and emitting offensive odors (Philippine Statistics Authority, 2017). Fish scales (FS) such as any other biomaterial are composed of organic and inorganic matter. Specific studies have established that the proteins, nitrogen-containing ligands (Yang, 1984), keratin, a protein material found in feathers, hair from mammals, and FS, with its sulfur groups (Coello, 1997), present in the organic fraction and other studies has been reported that hydroxyapatite or HAP, $Ca_{10}(PO_4)_6(OH)_2$, found and extracted from bones, sea shells, and FS present in the inorganic fraction (Sawa, 1996, Aiwaza, 1999) seem to be responsible for the adsorptive properties of FS. FS are a suitable alternative adsorption material, offering an adsorptive potential to a variety of dissolved material in water and moreover, they are readily available and at no cost since they are currently treated as a waste material, as they are inedible and is not a source of income for the fish farms (Srividya K, 2009).

Associated problems of highly dye-concentrated industrial wastewater and high cost treatment facilities have forced a new research to find alternative low cost biosorbent agents. It is in this light that the researchers have come up with the experimental study on the biosorption capability of utilizing the fish scale of Nile tilapia in the removal of color such as the Methylene Blue.

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The study aims to use *Oreochromis niloticus* (Nile Tilapia) fish scales (FS) to remove Methylene Blue dye in simulated wastewater. In the study, the FS must be pulverized and mixed with the dye solution.

Specifically, this study aims to fulfill the following specific objectives:

1. To determine the biosorption capacity of FS in removing dye in simulated wastewater made from:
   1.1 Distilled water
   1.2 Industrial wastewater
2. To determine the effects of the following varying conditions on the FS' removal efficiency of methylene blue for both simulated wastewater:
   2.1 Contact Time (15, 30, 60, 90, 120,150 and 180 minutes)
   2.2 Biosorbents Dosage (1g/L, 2g/L, 3g/L)
   2.3 Temperature (30□C, 50□C)
3. To use the Langmuir isotherm model to determine the adsorption pattern of methylene blue for both simulated wastewaters
4. To determine the maximum biosorption capacity of FS on Methylene Blue using percentage dye removal associate to the given varying condition

2. Materials and methods

The research study used experimental research design to obtain the feasibility of *Oreochromis niloticus* FS in removing color in simulated wastewater made from distilled water and industrial wastewater.

Figure 1 shows the step-by-step procedures of the whole experimental study. From the collection of FS of researchers to the production of biosorbents, sampling of raw industrial wastewater, preparation of the materials that were needed in the study and the preparation of simulated wastewater made from distilled water and industrial wastewater. Next was the batch of experiments for the treatment of the dye solutions. After those processes, the biosorption of the FS was analyzed for the biosorption capacity, removal efficiency and biosorption isotherm.

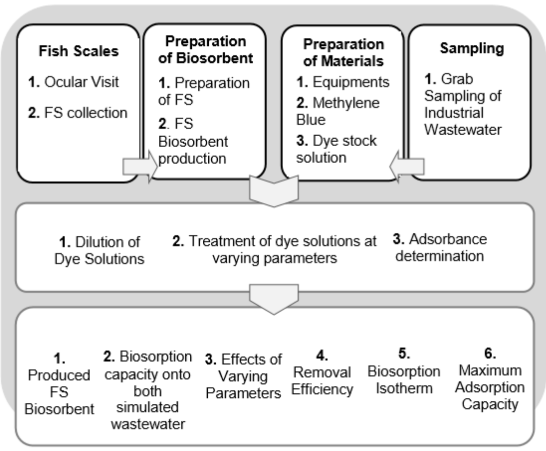

Figure 1. Research design.

The biosorbent used were the scales of fish *Oreochromis niloticus* (NileTilapia) that were collected from the local fish market at Batangas City.

The Methylene Blue was purchased at DKL Laboratory Supplies located at Espanya Street Sampaloc, Manila, Philippines. The equipment such as Water Bath with Shaker and UV/Vis Spectrophotometer of the Chemical and Food Engineering Laboratory at Batangas State University Main II Campus were used.

The industrial wastewater was gathered from an ice plant located at San Roque, Batangas. Ten (10) liters of wastewater samples were obtained on July 10, 2019 at 10:00 am to ensure a sufficient amount of wastewater for experiment.

3.1. Preparation of biosorbents

Raw fish scales (1.5 kg) were washed repeatedly to remove adhering dust, dirt and soluble impurities from their surfaces. Solids were separated by filtration and then ready for sun drying for 2 days. The biosorbent was transferred on a pan and dried in an oven at 60°C for 12 hours. The dried scales were then ground to fine powder using a Wiley mill grinder, and the pulverized scales were next sieved through 150 mesh to achieve 100 µm particle size and then used as a biosorbent without any pretreatment. One *Oreochromis niloticus* has the capacity to give approximately 0.01 grams of FS. Almost 150 *Oreochromis niloticus* were scaled to collect 1.5 kg of FS that can produce 200 grams of biosorbent. The amount of biosorbent produced may be increased depending on the grinding machine.

3.2. Preparation of dye solutions

The Methylene blue (DKL Laboratory Supplies) (color index: 52015; molecular formula: $C_{16}H_{18}N_3SCl$; molecular weight: 319.86, 662 nm) that was used in the study has commercial quality.

The dye stock solution (1,000 mg /L) was prepared by dissolving the required gram of dye in distilled water. The required concentrations of dye solutions were achieved by diluting the stock solution in a 250 ml Erlenmeyer flask with a suitable volume of distilled water and industrial wastewater.

3.3. Design of experiment

Batch biosorption experiments were performed at the laboratory to study the effect of the FS. Treatments had been repeated by changing the variables for the dye adsorption process in order to determine their effects on adsorption.

Table 1 shows the experimental design of the study. Twenty-one (21) controlled samples were prepared per setup. Seven (7) blank samples to cover the seven contact time variation was prepared for the whole experiment. The blank samples using distilled water are made similar with the controlled samples except for the addition of the biosorbent, subjected to the water bath at the same temperatures and contact time variations. Blank samples are also filtered prior to the reading of absorbance.

Table 1. Experimental design.

	T(°C)	Biosorbent dosage (g/100 ml)
DW	30	0.1, 0.2, 0.3
	50	0.1, 0.2, 0.3
IWW	30	0.1, 0.2, 0.3
	50	0.1, 0.2, 0.3

A total of four(4) set-ups incorporating the type of simulated wastewater, temperature, dosage and contact time as variables are made. Distilled and industrial simulated wastewater has two (2) set ups in each temperature used. The samples were subjected to the respective temperature (30□C, 50□C) in a water bath with shaker. In each set up, each biosorbent dosage (0.1, 0.2, and 0.3g/100 ml) has seven (7) samples, given that every time variation (15, 30, 60, 90,120, 150, and 180 minutes) one sample (readings are made triple times and the averaged was reported) was subjected to the UV/Vis for reading of absorbance. In total, there are 84 samples from the four set-ups with 21 samples each. Two trials are made per set-up comprising a total of 168 samples plus the 7 blank samples. The average between the two trials are reported and used in the analysis and data presentations.

The determination of percentage removal of Methylene blue in the simulated water was determined using the equation below:

$$R = \frac{(C_o - C_f)}{(C_o)} X\ 100\%$$

Where C_o = initial concentration of dye solution (mg/L) and C_f = final equilibrium concentration of dye solution (mg/L).

The linearized Langmuir equation is represented as follows [2]:

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{K_L q_m}$$

Where q_e is the adsorption capacity by weight at equilibrium, q_m is the monolayer adsorption capacity by weight, and K_L represents the equilibrium constant of adsorption reaction, and C_e is concentration of adsorbate at equilibrium.

The maximum biosorption capacity of FS was determined by using the equation below:

$$q_e = \frac{(C_o - C_e)V}{W}$$

where C_o is the initial concentration of dye solution (mg/L), C_e final equilibrium concentration of dye solution (mg/L), V (L) is the solution volume and W (g) is the adsorbent mass.

4. Results and discussion

4.1. Biosorption capacity of FS in the removal of methylene blue

Biosorption was a process of using waste biomass to remove contaminants on liquid mediums. The adsorption resulting in the interactions of dye and different varying conditions was monitored using UV-Vis Spectrophotometer at the respective maximum wavelength of methylene blue at 662 nm.

The biosorption capacities of FS powder in the removal of methylene blue for both simulated wastewaters were experimental set up was maintained similar in each simulated wastewater.

4.1.1 In simulated wastewater made from distilled water

The biosorption capacity of FS in simulated wastewater made from distilled water, as shown in Figure 2, was studied in dye solution with initial concentration of 10 mg/L, mixed with 0.2 g/100 ml biosorbent dosage at two different temperatures for a process time of 3 hours.

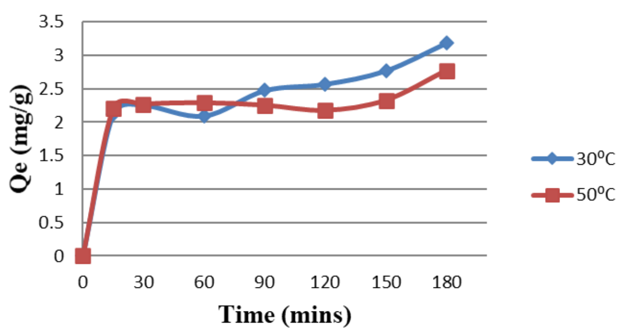

Figure 2. Biosorption capacity in simulated wastewater made from distilled water.

The biosorption capacity of FS increases with time and the sorption was initially fast in the first 15 minutes but then became slow as time increases. The temperature significantly affects the biosorption capacity of the FS that agrees to the findings of Uawonggul since the biosorption capacity was higher in lower temperature. In distilled water, there is an inverse relationship between temperature and biosorption capacity.

4.1.2. In simulated wastewater made from industrial wastewater

The biosorption capacity of FS in simulated wastewater made from industrial wastewater, as shown in Figure 3, was studied in dye solution with initial concentration of 10 mg/L, mixed with 0.2 g/100 ml biosorbent dosage at two different temperatures for a process time of 3 hours.

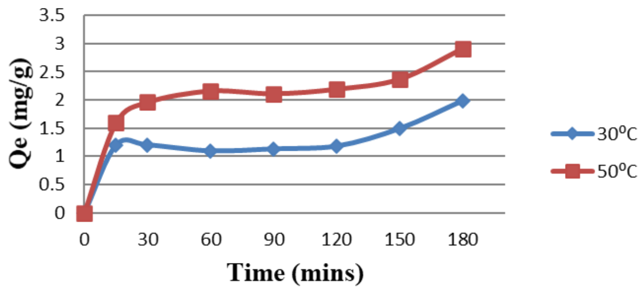

Figure 3. Biosorption capacity in simulated wastewater made from industrial wastewater.

The biosorption capacity increases with time and the rapid sorption takes place in the first 15 minutes but then a very small decrease in amount can be observed as time increases. The temperature directly affects the biosorption capacity of the FS given that the biosorption capacity was higher at 50□C, contrary to the result in distilled water which was shown in Figure 2.

4.2. Effects of the varying conditions on FS's removal efficiency of methylene blue

4.2.1. Effect of contact time for both simulated wastewaters

The effect of contact time on the adsorption of methylene blue dye for both simulated wastewaters by FS was carried out by mixing 0.2 g of biosorbent dosage to 100 ml dye solution at 30 and 50□C.

The effect of contact time was presented in Figure 4 at 30□C and Figure 5 at 50□C, which indicates that within 15 minutes the dye removal were 37.77% and 34.89 % for distilled water and 21.28% and 25.21% for industrial wastewater at 30□C and 50□C, respectively. The dosage of dye adsorbed, increases with the increasing contact time. The biosorption process was initially very fast due to the presence of binding sites on the biosorbent. However, the rate of removal gradually becomes slower as contact time increases.

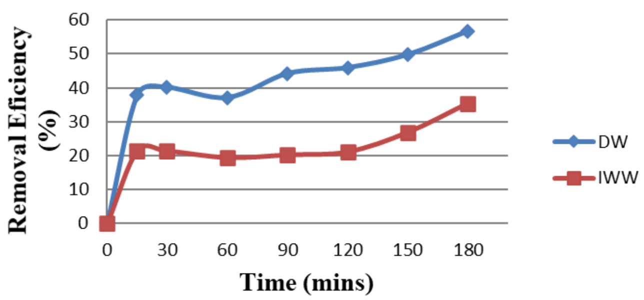

Figure 4. Effect of contact time on methylene blue dye removal efficiency at 30□C.

4.2.2 Effect of biosorbent dosage for both simulated wastewaters

The effect of biosorbent dosage was carried out by mixing varying dosage of FS powder in the solution ranges

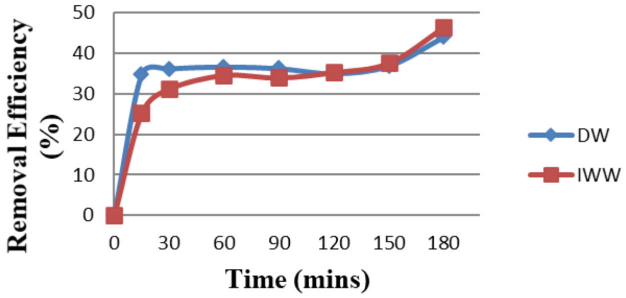

Figure 5. Effect of contact time on methylene blue dye removal Efficiency at 50□C.

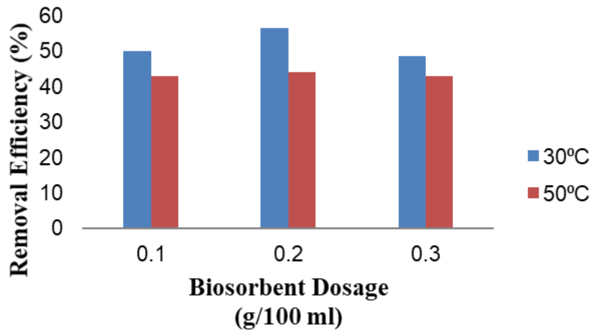

Figure 6. Effect of biosorbent dosage in simulated wastewater made from distilled water.

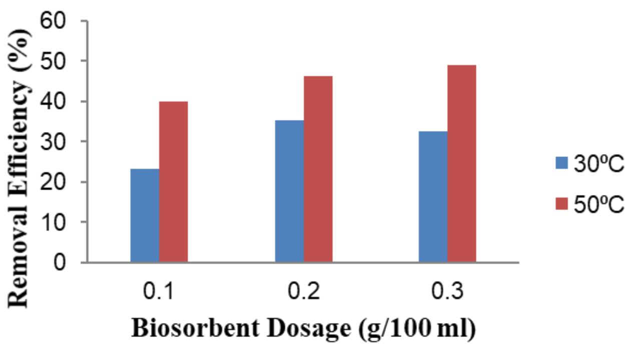

Figure 7. Effect of biosorbent dosage in simulated wastewater made from industrial wastewater.

from 0.1, 0.2 g and 0.3 g in a 100 ml dye solution at 30 and 50□C for 3 hours contact time. Figure 6 and Figure 7, show the effect of biosorbent dosage onto the removal efficiency of the FS. The percent removal was increased with an increase in biosorbent dosage for both simulated wastewater. The removal of dye in both simulated wastewater at 30 and 50□C temperature can be related to an increase in biosorbent surface area and availability of more biosorption or binding sites. However, the increase in biosorbent dosage led to decrease in removal of methylene blue especially to distilled water. The decrease in percentage dye removal may be due to saturation of remaining dye binding sites due to particulate interaction, which agrees to the result of Aksakal's findings where aggregation has led to decrease in total surface of the biosorbent.

4.2.3. Effect of Temperature for both simulated wastewaters

The biosorption of methylene blue on FS powder was studied by shaking and incubating the solutions using the water bath at two temperatures (30 and 50□C) for contact time of 3 hours.

In Figure 8, the effects of temperature onto the removal efficiency of FS for removing dyes from simulated

Figure 8. Effect of temperature on methylene blue dye removal efficiency.

wastewaters are different. The removal efficiency of FS had a large decrease in simulated wastewater made from distilled water when the temperature was at 50 □C. Uawonggul 2018 found that probably due to the configuration of FS that was broken and the amine groups would probably deviate from the FS while the temperature was increased. However, the removal efficiency of FS has a little increase when the temperature was at 50 □C in simulated wastewater made from industrial wastewater. According to the study of Chowdhury et al., the increase in temperature increases the mobility of the dye molecules and decreases the retarding forces acting on the molecules resulting in enhancement in the dye binding capacity of the biosorbent which is observed in the present study.

4.3. Biosorption isotherm study

The equilibrium biosorption isotherm was applied to understand the design and significance of adsorption systems. In general, it describes how adsorbates interact with adsorbents. Several isotherm equations were available and Langmuir biosorption isotherm was selected for this study.

The values of Langmuir constants for methylene blue dye uptake on FS were obtained in terms of equation (1) by using experimental adsorption results in this equation. Q_m and K_L were calculated from the slope and intercept of the straight lines of the plot $C_{eq}/\ q_{eq}$ versus C_{eq}. The related correlation coefficients ($\mathbf{R}^2$ values) are also summarized in Table 2.

As seen from Table 2, the Langmuir equation represents the adsorption process well. The $\mathbf{R}^2$ values were 0.988 and 0.978 in simulated wastewater made from distilled water and 0.988 and 0.964 in simulated wastewater made from industrial wastewater at 30 and 50□C, respectively, for both simulated wastewater indicating a good mathematical fit. The fact that the Langmuir isotherm fits the experimental data it assumes that distribution of binding sites onto the FS surface is homogeneous. The monolayer adsorption capacities for methylene blue onto FS at 30 and 50□C were found to be 1.186 and 0.962 mg/g in simulated wastewater made from distilled water and 0.396 and 0.750 mg/g in simulated wastewater made from industrial wastewater, respectively.

4.4. Maximum biosorption capacity of the FS

The highest equilibrium concentration, Q_e , was considered as the maximum Biosorption capacity. In Table 3 below is the summary of the maximum biosorption capacities and removal efficiency (%) of the methylene blue in each biosorbent dosage, temperature and simulated wastewater. Table 3 summarizes the maximum biosorption capacity and removal efficiency of methylene blue dye for each biosorbent dosage, temperature, and simulated water type. At

Table 2. Langmuir isotherm constants.

	Langmuir constant			
	T (□C)	q_m (mg/g)	K_L (L/mg)	R^2
DW	30	1.186	0.315	0.988
	50	0.962	0.218	0.978
IWW	30	0.396	0.170	0.988
	50	0.750	0.191	0.964

Table 3. Maximum biosorption capacity and removal efficiency of the FS powder.

	T(°C)	Biosorbent Dosage (g/100 ml)	Removal Efficiency (%)	Q_e mg of dye/ g adsorbent
DW	30	0.1	49.975	5.622
		0.2	56.631	3.185
		0.3	48.674	1.825
	50	0.1	42.910	5.401
		0.2	44.052	2.772
		0.3	43.042	1.806
IWW	30	0.1	23.355	2.627
		0.2	35.339	1.988
		0.3	32.539	1.220
	50	0.1	39.925	5.025
		0.2	46.335	2.916
		0.3	48.968	2.054

distilled water with 30 □C, the removal efficiency in percentage were 49.975, 56.631 and 48.674% which were higher than the removal efficiency at 50 □C, which was 42.910, 44.052 and 43.042% at 0.1g, 0.2g and 0.3g biosorbent dosage, respectively, for both temperature. Whereas to industrial wastewater with 30□C, the removal efficiencies were 23.355, 35.339 and 32.539%, which were slightly lower than the removal efficiency at 50 □C, which were 39.925, 46.335 and 48.968% at 0.1 g,0.2g and 0.3 g biosorbent dosage, respectively, for both temperature as well.

The plot between the biosorption capacity and removal efficiency at different temperatures and biosorbent dosage for simulated wastewater made from distilled water and industrial wastewater were shown in Figure 9 and Figure 10, respectively.

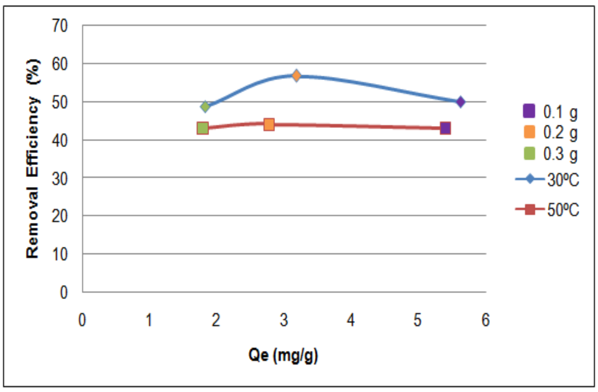

Figure 9. Plot between biosorption capacity and removal efficiency for distilled water.

Figure 9 shows the plot of biosorption capacity against the removal efficiency at varying biosorbent dosage (0.1, 0.2, and 0.3 g/100 ml) at two different temperatures (30 and 50 □C) for simulated wastewater made from distilled water. The removal efficiency was higher at 30□C in 0.2 g/100 ml of biosorbent dosage.

Figure 10 shows the plot of biosorption capacity against the removal efficiency at varying biosorbent dosage (0.1, 0.2, and 0.3 g/100 ml) at two different temperatures (30 and 50 □C) for simulated wastewater made from industrial wastewater. The removal efficiency was higher at 50□C in 0.3 g/100 ml of biosorbent dosage.

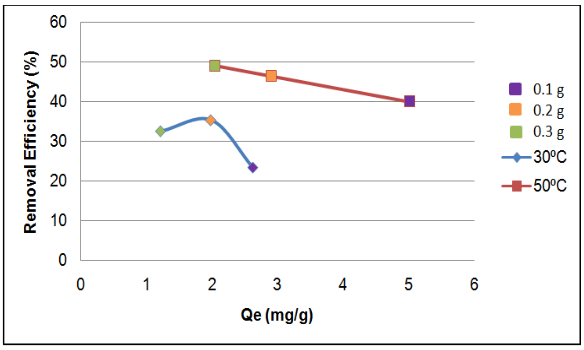

Figure 10. Plot of biosorption capacity and removal efficiency for industrial wastewater.

Table 3 and Figures 9 and 10 also show that the FS powder as biosorbent was much effective in simulated wastewater made from distilled water which incubated at 30□C and mixed with 0.2g/100 ml of biosorbent dosage.

Based from the findings and conclusions, the researchers hereby formulated the following recommendations:

1. A set up of 0.2 g per 100 ml dye solution with 10 mg/L concentration at 30□C of temperature is recommended to use since it give the highest removal efficiency among the experimental set up given.
2. The removal efficiency between the contact time of 0 to 15 minutes can be considered to see if there is significantt removal that happens within the span of time.
3. Future study is recommended on the use of even higher biosorbent dosages (> 0.3 g/100ml) when using industrial wastewater.
4. Future study is recommended on the use of *Oreochromis niloticus* fish scales as a biosorbent for the dye removal in simulated wastewater made from distilled water at lower temperatures and at higher temperatures for industrial wastewater to achieve higher removal efficiencies.
5. Exhibiting good biosorption capacity of Nile Tilapia fish scales as biosorbent on simulated wastewater made from distilled water and industrial wastewater, it is recommended to do further experiment on the adsorption capacity using the other types of wastewater such as kitchen wastewater, laundry wastewater, distillery wastewater, sugar industry wastewater and other industrial wastewater.
6. Utilization of the fish scales of other species that can easily be found as biosorbent and a comparative study of these is also recommended to be experimented for future study.
7. Utilization of other dye as an adsorbate is recommended to be experimented for future study to see if the same interaction will be observed.
8. Other biosorption isotherm models, like Freundlich isotherm model, can also be used to determine the adsorption pattern of FS in the removal of dye.

4. Conclusions

The present study shows that the low-cost fish scales of Nile Tilapia can be used as a biosorbent for the removal of methylene blue dye from simulated wastewater made from distilled water and industrial wastewater. The process parameters such as biosorbent dosage, contact time, and temperature are found to have a strong influence on the value of removal efficiency of FS. The biosorption capacity of FS powder as biosorbent varies on biosorbent dosage. FS, an inexpensive, easily available material and eco-friendly biodegradable bio-derived material, can be an alternative for more costly biosorbents used for dye removal in wastewater treatment processes.

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