Synthesis of Local Adsorbent from Azadirachta indica (Neem) for Bio-adsorption of Methylene Blue Dye from Aqueous Solutions

Zharama M. Llarena[1], Juvy P. Calibuso[2], Rosette M. Larua[3]

Pharmacy Department, School of Health Science Professions, St. Dominic College of Asia, Bacoor, Cavite, Philippines

Abstract— *Colored dye wastewater occurred as a direct consequence of dye production because of its widespread use in the textile and other commercial industries. There is a large production of dyes produced annually worldwide. These colorants were commonly used in textile, rubber, paper, plastic, and cosmetic industries. The conventional biological treatment process was ineffective in treating wastewater dyes due to its low biodegradation. It is often treated by either physical or chemical processes. However, these procedures were very expensive and could not be efficiently used to treat the wide range of waste dyes. This study generally aimed to determine whether local Azadirachta indica can bio-adsorb methylene blue dye. Specifically, this research aimed to compare the dye adsorption percentage of local Azadirachta indica and activated charcoal. It also aimed to determine whether different dosage strengths affect the dye removal. UV-Vis spectrophotometer was used to determine the absorbance of the dye samples treated with local Azadirachta indica and activated charcoal. Two-way ANOVA showed that there was no significant difference between Azadirachta indica and activated charcoal. Moreover, there was a significant difference in varying dosage strengths used. Hence, Azadirachta indica has a comparable bio-adsorption capacity with activated charcoal and its dosage strengths influence the dye adsorption.*

Keywords— adsorbent; bio-adsorption; neem; methylene blue.

I. INTRODUCTION

1.1. Background of the Study

Dyes present in our environment are deeply colored polymers and low in biodegradable activity [1]. Wastewater discharges from various industrial companies such as textiles, rubber, paper and plastics have several kinds of synthetic dyestuffs [2]. Dyes traced in commercial sewage create a serious harm to the environment [3]. In addition to that, releasing even little amounts of dye into water can greatly affect marine life and food chains due to the carcinogenic and mutagenic effects of synthetic dyes [2]. Methylene blue is an essential cationic dye. It is used in different textile industries, as it emits aromatic amines (e.g., benzidine, methylene), which are potential carcinogens [4]. It is often used in cotton, wool, and silk dyeing. MB has numerous effects in humans and animals, namely, eye burns, methemoglobinemia, cyanosis, convulsions, tachycardia, dyspnea, irritation to the skin, and if swallowed, irritation to the gastrointestinal tract, nausea, vomiting, and diarrhea [5]. The methylene blue removal from any wastewater is of primary importance because of its serious environmental destruction that can happen as a consequence of contact with it, specifically in the case of people [6].

Bio-adsorption has been used in the removal of color from discharges. Activated carbon is the widely used adsorbent [7]. Due to its expensiveness and considering the large quantity of discharge produced by textile manufacturers, researches are turning toward the use of alternative adsorbents, also called non-conventional low-cost adsorbents. Plastic, paper, textile and cosmetics industries use dyes to color their products. These dyes are widespread water pollutants and they may be commonly found in detectable amounts in commercial waste water. Their existence in water, even at very low concentrations, is greatly visible and unwanted. Adsorption has been used in commercial procedures for separation and purification. In wastewater treatment, activated carbon has long been used as a standard adsorbent for dye removal. In spite of its worldwide use in various cleaning processes, activated carbon remains at high cost; therefore, the development of cheap alternative adsorbents has been the focus of recent research. Contributions in this concern have been done by many researchers who have used a number of materials such as agricultural wastes: coir pith, banana pith, sugar cane dust, sawdust, activated carbon fibers and rice hulls,

industrial solid wastes: fly ash, red mud and shale oil ash, and so on [8].

In general, this study aimed to determine whether local *Azadirachta indica* can bio-adsorb methylene blue dye from aqueous solutions. Specifically, this research aimed to compare the dye adsorption percentage between of local *Azadirachta indica* and activated charcoal. It also aimed to determine whether different dosage strengths affect the dye removal.

II. MATERIALS AND METHODS

2.1 Research Design

This study followed an experimental design. *Azadirachta indica* and activated charcoal were used as intervention groups to reduce the dye content present in varying strengths of methylene blue dye aqueous solutions. Hitachi UV-Vis Spectrophotometer was used to measure the absorbance before and after treatment protocols.

2.2 Collection and Preparation of Sample

Azadirachta indica was collected from DOST Science Garden. The leaves were washed repeatedly by using distilled water to remove moisture and soluble impurities. The leaves were dried using direct sunlight for removal of moisture. The dried leaves were crushed and kept in an oven for 3 hours at 250°C [9]. The sample was further ground to obtain a black powder and weighed separately at 0.25g, 0.5g, 1.0g, 1.5g and 2.0g.

2.3 Preparation of Methylene Blue Dye Aqueous Solutions

0.5g of methylene blue dye was weighed and dissolved in 1 L of distilled water. An aliquot of 20 mL was further diluted with 30 mL distilled water to make 50 mL of 40% solution. Another aliquot of 30 mL was diluted with 20 mL distilled water to make 50 mL of 60% solution. The last was an aliquot of 40 mL further diluted with 10 mL distilled water to make 50 mL of 80% solution.

2.4 Adsorption of Methylene Blue Dye

Set A, B and C contained varying strengths of 40%, 60% and 80% methylene blue dye aqueous solutions. Each set was treated with different dosage strengths of *Azadirachta indica* powder and activated charcoal. Set A was treated with 0.25g, 0.5g and 1.0g of *Azadirachta indica* powder and activated charcoal. Set B was treated with 0.5g, 1.0g and 1.5g, while Set C was treated with 1.0g, 1.5g and 2.0g of *Azadirachta indica* powder and activated charcoal. After 30 minutes of treatments, all samples were filtered and placed in PET bottles.

2.5 UV-Vis Absorbance Determination

An aliquot of 100 µL was dissolved in 2 mL distilled water. Dye concentrations were determined by using absorbance values measured before and after the treatments. The λ max was set at 650 nm with Hitachi UV Visible Spectrophotometer [10].

III. RESULTS AND DISCUSSION

Azadirachta indica and activated charcoal shows to lower the absorbance of methylene blue dye aqueous solutions (Table 1). Varying dosage strengths has lowered the concencentrations of methylene blue dye. The dye adsorption percentage of *Azadirachta indica* ranging from 70.31% - 97.24% was compared with the results of dye removal capacity of activated charcoal ranging from 69.40% - 97.45% (Table 1).

Table 1. Dye Absorbance and Dye Adsorption % of Samples In Varying Strengths

Sample Name	Absorbance	Dye Adsorption %
MB 40%	1.374	
MB 60%	2.229	
MB 80%	3.954	
Set A1	0.408	70.31%
Set A2	0.545	75.55%
Set A3	0.654	83.46%
SetB1	0.182	86.75%
SetB2	0.224	89.95%
SetB3	0.208	94.74%
SetC1	0.093	93.23%
SetC2	0.096	95.69%
SetC3	0.109	97.24%
SetD1	0.340	75.25%
SetD2	0.682	69.40%
SetD3	0.753	80.96%

SetE1	0.196	85.74%
SetE2	0.190	91.48%
SetE3	0.275	93.05%
SetF1	0.092	93.30%
SetF2	0.098	95.60%
SetF3	0.101	97.45%

Two-way ANOVA illustrates that there was no significant difference between *Azadirachta indica* and activated charcoal. Moreover, there was a significant difference in varying dosage strengths used (Table 2). Critical regions were set at f1>4.75 and f2>3.89.

Table 2. Two-way ANOVA Analysis

ANOVA Summary	2rows x 3columns			
Source	SS	df	MS	F
Rows	1.22	1	1.22	0.06
Columns	1239.14	2	619.57	31.87
r x c	1.31	2	0.66	0.03
Error	233.25	12	19.44	
Total	1474.92	17		

IV. CONCLUSIONS

Dyes accumulate in industrial effluents. It can sequester metal ions and carcinogenic in nature, thus, causing harm to many people. Based on the results of the study, *Azadirachta indica* has a comparable bio-adsorption capacity with activated charcoal and its dosage strengths influence the dye adsorption. Hence, *Azadirachtaindica*can be a cheaper alternative for low cost bio-adsorbent.

V. RECOMMENDATIONS

Time, temperature and pH kinetics are recommended for the further study of bio-adsorption of *Azadirachta indica* powder. Biosorption of heavy metals are also suggested as well. Furthermore, more low cost bio-adsorbents are recommended to be studied.

REFERENCES

[1] Khatod, Indira (2013): Removal Of Methylene Blue Dye From Aqueous Solutions By Neem Leaf And Orange Peel Powder, International Journal of ChemTech Research, Vol.5, No.2, pp 572-577

[2] PrasanaManikandaKartik, J, et al (2012): Removal of Direct Red Dye Using Neem Leaf Litter, Helix, Vol. 1(2):129-133

[3] Sunil Kumar, V, et al (2012): Removal of Methylene Blue from Aqueous Effluent Using Fixed Bed of Groundnut Shell Powder, Journal of Chemistry, Volume 2013, 5 pages

[4] Srinivasa Reddy, B, et al. (2012): Removal of methylene blue dye from waste waters using new bio-sorbents derived from *Annonasquamosa and Azadiractaindica*plants, Journal of Chemical and Pharmaceutical Research, 2012, 4(11):4682-4694

[5] Ali Fil, B, et al (2012): Cationic Dye (Methylene Blue) Removal from Aqueous Solution by Montmorillonite, Bull. Korean Chem. Soc. 2012, Vol. 33, No. 10

[6] Wong, YC, et al (2013): Removal of Methylene Blue and Malachite Green Dye Using Different Form of Coconut Fibre as Absorbent, Journal of Basic & Applied Sciences, 2013, 9, 172-177

[7] Oboh, Innocent, et al (2013): Biosorption of Heavy Metal Ions from Aqueous Solutions Using a Biomaterial, Leonardo Journal of Sciences, Issue 14

[8] Pandhare, Ghanshyam, et al (2013): Synthesis of Low Cost Adsorbent from *Azadirachta indica* (Neem) Leaf Powder, International Journal of Advanced Engineering Research and Studies, IJAERS Vol. II

[9] Pandhram, Parineeta, et al (2013): Adsorption of Chromium from Industrial Waste Water By Using Neem Leaves as a Low Cost Adsorbent, International Journal of Chemical and Physical Sciences, IJCPS Vol. 2

[10] Velmurugan, P, et al (2011): Dye removal from aqueous solution using low cost adsorbent, International Journal of Environmental Sciences, Volume 1, No 7.