

Activated carbon adsorbent derived from *Erythrina brucei* stem powder on industrial waste water treatment

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Abstract— A wide range of adverse environmental toxic agents were adulterated in the Industrial waste-water. One of the major pollutant in Industrial effluent is dye materials, even a small amount of dye can alter the quality of water source, that can cause harmful impacts on living organism. Therefore, it is paramount to eradicate the dye from the industrial effluents. Methylene Blue (MB) is a commonly used dye in various industries that has a number of negative impacts on humans and animals. The aim of this investigation is to study the potential of activated carbon (AC) prepared from *Erythrina brucei* stem powder (EBS) adsorbent for the treatment of methylene blue containing wastewater. To determine the optimum condition to attain the maximum removal efficiency, the effect of adsorbent dose, initial concentration, pH of the solution and contact time of AC prepared from EBS were investigated. The absorption spectroscopy was used to monitor the adsorption potential of the adsorbent. The experimental results showed that under optimum condition (0.3g/100ml adsorbent dose with 60 min contact time at pH 3) 99.97 % of the MB was adsorbed by the activated carbon prepared from EBS. The result suggests that AC prepared from EBS could be employed as low cost alternatives to commercial activated carbon in wastewater treatment for the removal of basic dyes. This effective and low cost removal method may provide a promising solution for the removal of methylene blue dye from wastewater.

Index Terms— *Erythrina brucei* stem, Activated carbon, Methylene blue, Waste water, Adsorption process.

1 INTRODUCTION

Water is pivotal for the existence of all living organisms on earth. Worldwide, adulteration of fresh water sources with industrial effluents has been a key anxiety. Among the various contaminants, dyes have been the major class of pollutant that often present in industrial waste water. In general, dyes are coloring agents, which has been used in a range of industries mainly in textiles, leather, printing, food, and plastics. The dye-containing industrial wastewater often cause severe environmental complications. Specifically, the dyes prevent the penetration of light that affect the photosynthesis of water streams and interrupt the aquatic equilibrium. It has been reported that even a trace amount of dyes (<1ppm) presence in water can cause harmful health effect in humans such as mutagenic and carcinogenic [1]. Therefore, removal of dyes from industrial wastewater before discharge into water stream is paramount to preserve the water quality.

Methylene Blue (MB), is an industrially well celebrated cationic dye [2], is universally used as the coloring agent in textile industry. The major concern in MB is carcinogenic and very hard to decompose. In addition to that MB dye has various harmful effect on humans and animals that includes quadriplegia, esophagus, and diarrhea. Thus, removal of MB dye from wastewater is essential for humans and protect the environment [2].

Recently, several techniques have been explored for the removal of dye from industrial waste-water such as flocculation, adsorption, oxidation, electrolysis, biodegradation, ion-exchange, photo catalysis [3, 4]. Adsorption technique has been found to be a superior separation and purification method to other methods due to its easy nature, low cost, high selectivity, and high efficiency [13, 5-6]. Among the various adsorbents, activated carbon is the most preferred adsorbent for the removal of dyes because of its excellent adsorption ability [7, 9, 10]. However, widespread use of activated carbon is restricted because of

its high cost and source [8].

Erythrina brucei, is an indigenous tree found only in Ethiopia, and, is grown in every highlands of Ethiopia and very cheap (Fig. 1). Traditionally, it has been used for Firewood, carving (bee-hives and drums), medicine (bark and roots), nitrogen fixation, and soil conservation. Therefore, production of activated carbon from this plant for large scale application is very promising in respect to the raw material abundance and ease of its preparation. In addition, the stem part of the *Erythrina brucei*



Fig. 1. Plants raised from rooted cuttings.

has not been used in wooden industry. To our knowledge and the literature survey reveals that so far no considerable effort has been made to study the treatment of MB dye by activated carbon prepared from *Erythrina brucei* stem (EBS). Therefore, in this research the efficiency of AC prepared from EBS as an adsorbent will be investigated for removing MB, dye from wastewater using the adsorption process.

2 MATERIALS AND METHODS

The EBS was collected from bekoji, an Arsi zone of Oromia region in Ethiopia. All chemicals used were analytical reagent grade. Methylene blue (MB) (MW319.859 g/mol basic blue), dried and distilled water was used for preparation of stock solution. HCl and NaOH were used to adjust the pH of the solution. Distilled water was used for experimental study.

2.1. Preparation of activated carbon from Erythrina bruceias adsorbent.

The stem of collected Erythrinabyuceisamples were frombekojidried in sun light until they become moisture free. Ebs sample were cut into small pieces (0.5 - 1.0 cm in length), washed with distilled water and oven dried at 120 °C overnight. Then the dried samples were ground using domestic grinder and then were sieved using standard sieve with matrix size of 75-300 µm to obtain fine powder of uniform particle size. Then transferring the sample powder to 250 ml beaker, and was soaked with sufficient amount of concentrated HCl and left overnight at room temperature in order to: extract the compound formed and reagent excess and degrade or dehydrate the organic molecules during carbonization that prevent deposition of hydrocarbon on the carbon surface. The acid treated Erythrina brucei of powder was then washed thoroughly with distilled water and then filtered with Whatman number one filter paper. The residue was transferred into a clean beaker, to which enough amount of 0.1M NaOH solution was added and soaked again overnight so that any excess acid present was neutralized. The treated solid residue was washed again with distilled water till attaining a constant pH and then made to dry at room temperature. The powder obtained was kept in a hot air Oven at 100 °C for a day. The impregnated precursor was carbonized in oven at 225 °C for 1h. [38-39], then the remaining EBS (activated carbon) was cool and stored in desiccators for later used for adsorption studies. Carbonization, the conversion of organic matter to elemental carbon at high temperature in an inert atmosphere is the first stage in the physical activation process. This results in the elimination of elemental hydrogen and oxygen in the precursor to produce a carbon skeleton possessing a latent pore structure. [29, 30] Typical temperatures for the carbonization step ranges from 300 °C to 500 °C [31].

2.2. Determination of Physiochemical properties of the EBS adsorbent.

2.2.1. Determination of the moisture content of the adsorbent.

One gram of adsorbent was weighed accurately in an evaporating dish and placed in an electric oven at 110 °C for 2 hr. it was then cooled and weighed again. The process of heating, cooling and weighing of the material was repeated several times at 30 min intervals until the difference between two consecutive weighing become less than 0.01g [32]. 0.01g indicates how much the weighed adsorbents are near to each other. The moisture content will be determined using equation 10 below:

$$\text{Moisture content by weight} = \frac{M-X}{M} \times 100 \dots\dots\dots (10)$$

Where M is weight of adsorbent taken for the test (g) and X is weight of adsorbent after drying both in grams.

2.1.2. Determination of the ash content

One gram of adsorbent was weighed accurately in a porcelain crucible and placed in an electric oven at 110 °C for 2 hr. The adsorbent kept in a porcelain crucible in an electric oven was then removed from the oven & heated in a muffle furnace at a temperature of 500 °C for 2 hours. The crucible with its content was removed from the furnace and cooled to room temperature and weighed again. The process of heating, cooling and weighing of the material was repeated several times at 30 min intervals until the difference between two consecutive weighing become less than 0.01g [40]. The ash content was determined using Equation 11 below:

$$\text{Ash content percent by weight} = \frac{M1}{M2-X} \times 100 \dots\dots\dots (11)$$

Where M1 is weight of the ash (g),
 M2 is weight of adsorbent will be taken for the test (g) and
 X is weight of adsorbent after drying both in grams moisture content present in the material.

2.1.3. Preparation of adsorbate (Stock Solution preparation).

Preparations of stock solution of methylene blue was carried out by dissolving 1gm of (MB) in1000 ml of distilled water in order to get 1000 ppm concentration, while the working concentrations were prepared by using the equation., equation 12 .

$$C1 V1 = C2 V2 \dots\dots\dots (12)$$

The concentration of MB was measured using UV-visible spectrometer. A calibration curve isPlot between absorbance and concentration of dye solution to obtain absorbance concentration Profile.

2.3. Adsorption Studies

The experiments on the adsorption of MB, on AC prepared from EBS were conducting out in reciprocating shaker at 100 rpm using 100mL of flasks an aqueous solution of the dye in concentration of 25mg/L , 50 mg/L, 75 mg/L, and 100 mg/L. To the resulting solution, 300mg of adsorbent was added to each 100 ml flask at a neutral pH and room temperature. The initial pH value of each solution was adjusted with 0.1M HCl or 0.1M NaOH using a pH meter. In all experiments the flasks were sealed to prevent any change in volume during the experiment. After shaking the flasks after equilibration time, the adsorbents and the supernatant solution were separated by filtration. After filtration, the concentration of MB, in the supernatant solution was analyzed using an UV-visible spectrophotometer by recording the absorbance changes at a wavelength of maximum absorbance (665nm). The percentage removal of dye (ad-

sorption) and amount of dye adsorbed on adsorbents at equilibrium was calculated by equation (13) and (14) respectively.

$$\text{Removal efficiency (\%)} = \frac{[C_0 - C_e]}{C_0} \times 100 \dots \dots \dots (13)$$

$$q_e = \frac{[C_0 - C_e] V}{M} \dots \dots \dots (14)$$

Where, C₀ = Concentration of MB, dye in the sample solution before treatment

C_e = Concentration of MB, dye in the sample solution after treatment

q_e = amount of MB, dye adsorbed on adsorbents at equilibrium.

V = volume of MB solution

M = mass of adsorbent (Ebs) The effects of various parameters such as pH, contact time, MB, dye concentration, adsorbent dosage, ionic strength and temperature on adsorption of MB onto activated carbon prepared from Ebs was study.

2.4. Effects of Various Parameters on the Adsorption of MB on the Activated Carbon of EBS.

2.4.1. Effect of initial dye concentration

To investigate the effect of initial dye concentration on 300 mg of AC prepared from EBS was added to different MB dye concentrations (25 ppm, 50 ppm, 100 ppm, 150 ppm and 200 ppm) which was prepared from the dye stock solution in 100 ml flasks. All of the samples were agitated on shaker for 1h. The adsorbent and supernatant was separated by filtration. Finally, the concentration of MB, solution free from any suspended carbon will be estimate by measuring its absorbance at 665 nm using UV-Vis spectrophotometer by kept other parameters constant.

2.4.2. Effect of adsorbent dosages

To determine the effect of adsorbent dosage 25 ppm MB, dye concentration was added to different AC prepared from EBS doses (50, 100, 200 and 300 mg) in a 100mL flask and agitated under mechanical shaker for 1h. The adsorbent and supernatant was separated by filtration. Finally, the concentration of MB, solution free from any suspended carbon will be estimate by measuring its absorbance at 665 nm using UV-Vis spectrophotometer by kept other parameters constant.

2.4.3. The effect of contact time.

The effect of contact time was tested on the potential of AC prepared from EBS by mixing 300mg of the AC prepared from EBS with different initial concentration (25, 50, 75 and 100mg/L) of MB, solution in 100 mL flask for 20 - 80 min. 5 ml of this solution (sample) was withdrawn at 10 min interval from the flask and the absorbance was measured using UV-VIS at 664 nm by keeping other parameter constant Finally the amount of MB, adsorbed and percent removal of MB, dye was calculated and the plot of percent removal of MB, dye as well

as the amount of MB, dye adsorbed was plotted.

2.4.4. Effect of solution pH

The influence of solution pH (2-8) was examined to understand the adsorption mechanism. 100ml of the dye solution was mixed with constant concentration of AC prepared from EBS (300mg) at 100 rpm in a 100ml flask and agitated under mechanical shaker. The adsorbent and supernatant was separated by filtration. And the absorbance was measured using UV-VIS at 665nm.

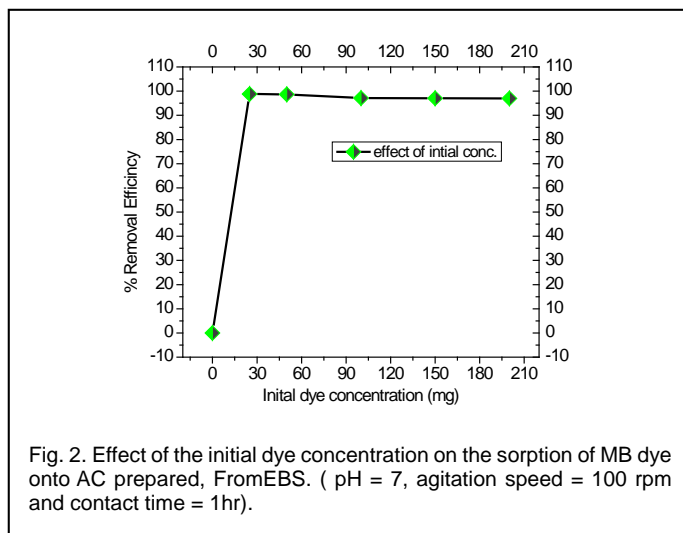
3. RESULTS AND DISCUSSION

This investigation aims to study the potential of activated carbon prepared from EBS. The laboratory investigations made for EBS characterization and study of effects of different parameters such as pH, concentration of methylene blue and contact time on MB removal capacity of EBS from constituent synthetic water are presented. The constituent wastewater loaded with MB dye was treated with EBS and performance was evaluated by varying the parameters to optimize those factors which affect the performance of EBS for specific dye.

3.1. Optimum Conditions for Methylene Blue Removal by the Erythrina brucei Stem Adsorbent

3.1.1. Effect of Initial Concentration on MB Adsorption

The effect of initial concentration on the adsorption of MB on AC prepared from EBS was studied in the range (25-100 mg/L). The result presented in (Figure 2) indicates that after determination of moisture of AC prepared from EBS. When 300 mg of



AC prepared from EBS was added to different MB dye concentrations (25 ppm, 50 ppm, 100 ppm, 150 ppm and 200 ppm) in 100 ml flasks and agitated all samples on shaker for 60 minutes and after filtration, withdrawn 5 ml of sample from the flask by measured the absorbance using UV-VIS at 665 nm, the

percentage removal of MB dye decreased from 98.812 % to 97.01% with increasing in initial concentration of MB dye due to the fixed total number of adsorbent available sites, given adsorbent dose adsorbing almost the equal amount of adsorbate, which resulting in a decrease in the removal of the adsorbate . While the amount of MB adsorbed increases from 8.23 mg/g to 65.67 mg/g with increase of dye initial concentration) due to the increased ratio of number of moles of MB dye to the vacant available sites. (Appendix Table 1.1). For a given adsorbent dose the number of adsorbent available sites was fixed thus adsorbing almost the equal amount of adsorbate, which resulting in a decrease in the removal of the adsorbate, consequent to an increase in initial MB concentration [22]. Therefore, it was evident from the results that MB adsorption was dependent on the initial MB concentration.

3.1.2. Effect of adsorbent dose

The results, of adsorbent dose study are given in Figure (3). The removal efficiency increases from 98.24 % to 99.6% with an increase in adsorbent dose from 0.05 to 0.3g/100ml. This can be attributed to increased number of sorption sites available for sorbent solute interaction. With further increase of adsorbent dose from 1.0 to 2.0 g, the percent adsorption increases less significantly. Thus, the adsorbent dose was maintained at 0.3 g in all the subsequent experiments, which was considered to be sufficient for the removal of MB. According to, [24] increase in adsorption with adsorbent dose can be attributed to increased adsorbent surface area and availability of more adsorption sites. In some case the amount of MB adsorbed per unit weight of adsorbent decreases with the adsorbent dose. This is due to the fact that at higher adsorbent dose the solution ion concentration drops to a lower value and the system reaches equilibrium at lower values of indicating the adsorption sites remain unsaturated [32].

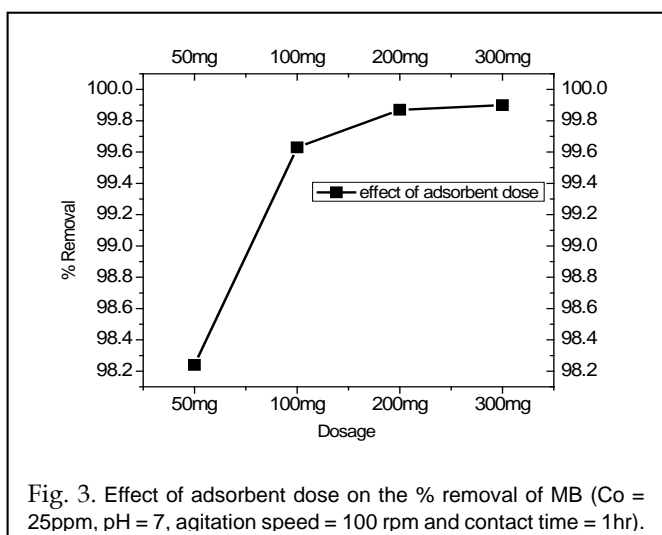


Fig. 3. Effect of adsorbent dose on the % removal of MB (Co = 25ppm, pH = 7, agitation speed = 100 rpm and contact time = 1hr).

3.1.3. Effect of pH

pH is a vital parameter in adsorption studies. Variations of pH can control the uptake of MB from wastewater and aqueous solutions. Figure 4 show the effect of pH on MB removal efficiencies of EBS adsorbent. The studies were conducted at different pH with an initial MB concentration of 25 ppm in 100ml solution, using constant adsorbent dose 0.3 g/100ml solution and agitation period of 60 minutes for MB. The results shown that the percentage of adsorption increases with pH. The maximum adsorption was achieved at pH 3 and thereafter it decreases with further increase in pH. Maximum removal efficiency at optimum pH is about 99.97% MB from solution with initial MB concentration of 25ppm. This may be because at pH values below 4.0, the electrostatic force of repulsion between adsorbent (EBS) and adsorbate (MB) is prominent. At pH above 6.0, there is a possibility of adsorbate precipitation on the surface of the adsorbents by nucleation (In Fig 4) higher pH, that is, above optimum pH of 3, increase in OH⁻ ions cause a decrease in adsorption of MB at adsorbent adsorbate interface. Lower solubility's of hydrolyzed MB species may be another reason for the maximum adsorption at 3 pH. Since, in lower pH range, MB is present predominantly as MB in the adsorptive solution, there is a competition between H⁺ and MB⁺ ions for adsorption at the ion exchangeable sites, leading to a low removal of MB. The extensive repulsion of MB due to protonation of the adsorbent surface at higher pH may be another reason for decrease in adsorption of MB in higher pH range [38].

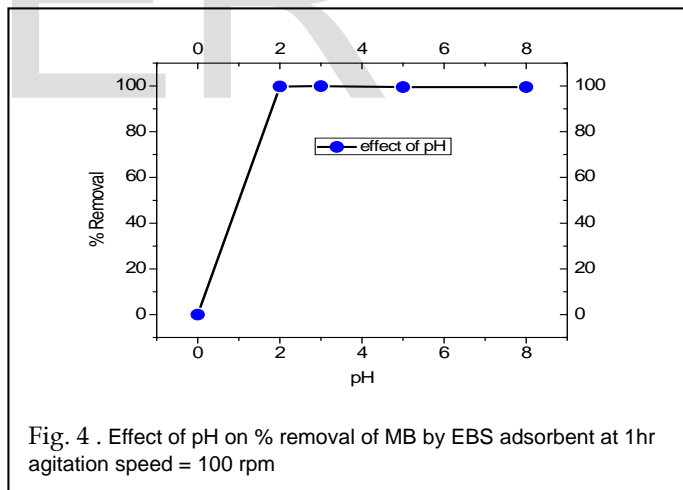


Fig. 4 . Effect of pH on % removal of MB by EBS adsorbent at 1hr agitation speed = 100 rpm

3.1.4. Effect of contact time.

The variation in the percentage removal of MB with contact time using 0.3g/100ml of EBS adsorbent at 7pH for varying initial MB concentration of 25ppm, 50ppm and 75ppm was shown in Fig 5 maximum percent removal of MB was 99.66% at 60 minutes and concentration of 25ppm in each case. This was achieved by varying the contact time from 20min to 80 min. in separate experimental runs. As expected, the amount of MB adsorbed into adsorbent increases with time, and at some point, reaches a constant value beyond which no more is removed from solution. At this point, the amount of the MB desorbing from the adsorbent is in a state of dynamic equilibrium with the

amount being absorbed to the adsorbent. The time required to attain this state of equilibrium is termed the equilibrium time, and the amount of MB adsorbed at the equilibrium time reflects the maximum adsorption capacity of the adsorbent under those operating conditions. Equilibrium was reached at 60 min. in all cases. This can be explained by the fact that initially, the rate of MB uptake was higher because all sites on the adsorbent were vacant and MB concentration was high, but decrease of adsorption sites reduced the uptake rate. Similar results were found in the study of [37].

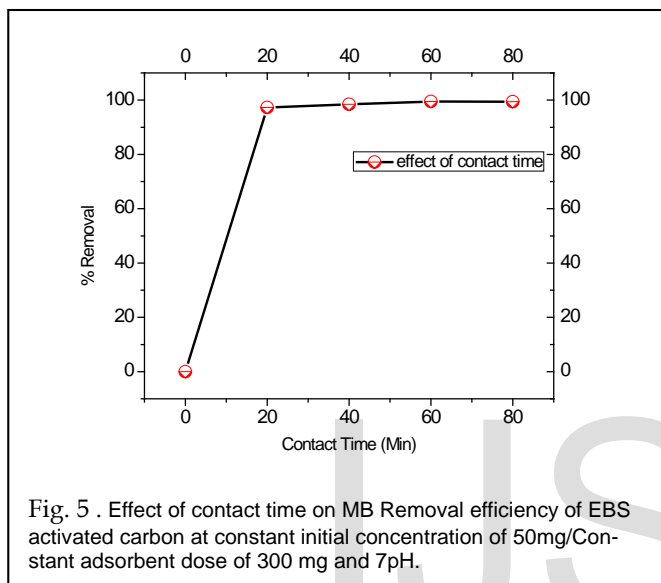


Fig. 5 . Effect of contact time on MB Removal efficiency of EBS activated carbon at constant initial concentration of 50mg/Constant adsorbent dose of 300 mg and 7pH.

4. CONCLUSION

In this study, low cost adsorbents were successfully prepared from *Erythrina brucei* stem. The results indicate that the AC prepared from EBS might be used as an adsorbent for the removal of MB dye from wastewater. Further, the adsorption of recalcitrant compounds from synthetic wastewater using these locally available low cost adsorbents was studied. Additionally, response surface methodology was used to investigate the interactive effect of the operating parameters namely; adsorbent dose, initial concentration, contact time and solution pH. EBS may be low cost and effective adsorbent for the removal of MB from waste water. Experiment results showed that maximum removal of MB by EBS at optimum condition (3 pH, 60 min. contact time, 0.3g/100ml adsorbent dose and 25ppm concentration) is 99.85%. These experimental studies on adsorbents would be quite useful in developing an appropriate technology for the removal of organic dye from contaminated industrial effluents. Overall, the result indicates that the EBS adsorbent is capable of removing MB dye from aqueous solution. Therefore, EBS can be used, as a low cost and abundant source, for the removal of MB and it may be an alternative to costly materials. Further research on the ability of EBS adsorbent is sought for its simultaneous uptake of MB along with other toxic heavy metals such as Cd, Pb, Hg, Cu and Zn may pave the way for in-situ heavy metal removal.

Conflicts of Interest

The authors declare that they have no conflicts of interest.

Authors' Contributions

The experiments and the first draft writing were conducted by Abdulfeta Muhdin, Biruktawit Tefaye, and Abdulrazak Heiredin, and the supervision and edition were done by Prof. Rajalakshmanan Eswaramoorthy. The Co-authors Bedasa Abdisa Gonfa, Fedlu Kedir Sabir were participated in analyzing the results and drafting the manuscript.

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