Adsorption of Methylene Blue dye using Marine algae *Ulva lactuca*

D Pratiwi, D J Prasetyo, C D Poeloengasih

Research Unit for Natural Product Technology, Indonesian Institute of Science, Yogyakarta, Indonesia 55861

Corresponding author: dee.diahpratiwi@gmail.com

**Abstract.** Contamination of dyes causes an aesthetic problem, severe public health concern and adds several serious environmental issues as a result of their resistance in nature and nonbiodegradable characteristic. Adsorption of methylene blue (MB) has been conducted by utilizing marine algae *U. lactuca* as biosorbent. In this study, the batch adsorption of Methylene blue dye using marine algae *U. lactuca* was examined. The ability of the *U. lactuca* in removing the dye color was dependent on contact time, algae biomass, dye concentration, and pH. The optimum adsorption was found at around pH 8; contact time 110 min; adsorbent dose 1.25 g/L; initial concentration 25 mg/L. The maximum percentage dye removal value was 91.92%. This work confirms the potential use of *U. lactuca* for removing Methylene blue dyes solutions.

**Keywords:** Methylene blue, *Ulva lactuca*, adsorption, biosorbent, removal of dyes

1. **Introduction**

A dye is a substance that used to impart color to product or material i.e. paper, leather, textile, rubber, plastic, and cosmetic. Colour added to the product or material to create an attractive appearance and give added value to an aesthetic and also for a social reason. Coloring compounds can be made from a natural compound or human-made (synthetic). Synthetic dyes are aromatic water soluble and dispersible organic colorants [1,2]. The dyes are having potential application in various industry and have become a major commodity in trade.

The textile industry in one of the major application for synthetic dyes. These industries produce colored wastewater which is heavily polluted with dyes, textile auxiliaries, chemicals used in the process production [3]. Visual pollution is a major problem in water quality because it is not easy to accept red or brown rivers [4]. Contamination of dyes causes not only an aesthetic problem and severe public health concern but also adds several serious environmental issues as a result of their resistance in nature and nonbiodegradable characteristics [5-6]. The textile industrial wastewater could lead to a decline in the quality of the environment. The level of pollutant even in very low concentration is highly visible, undesirable, carcinogenic and will affect aquatic life and food chain. They are generally stable to light, an oxidizing agent and resistant to aerobic digestion, which is raising difficulties in treating colored wastewater and difficult to be degraded.

Colour can be removed from wastewater effluent by chemical, physical, and biology methods [7]. Some of the applied methods in the treatment of effluent containing dyes are flocculation, coagulation, precipitation, adsorption, filtration, and ion exchange. All these methods have shortcomings and
limitations, sometimes the process is not always effective and economical [1,8]. This problem has prompted the employment of various material to develop a cheaper alternative method by utilizing many varieties of biosorbent which has high sorption capability and an unlimited amount of availability. Marine alga is an alternative adsorbent from biological resources which are obtainable in massive quantities in the world. Bioconversion and biosorption could utilize by using marine algae as an adsorbent. Through bioconversion, some algae will break down the dyes into additional easy compounds. Biosorption process is typically fast and in theory appropriate for extraction of color and metal ion from massive volumes of water. Hence, biosorption is thought of as a promising technique involved with the uptake of unwanted ions from liquid solutions using biological materials [2,8].

*U. lactuca* is a small genus of marine algae which are available in large quantities in the world. This marine alga is an edible green alga as known by the common name sea lettuce. *U. Lactuca* was approved as usable biomaterials for biological treatment for dye effluent [2,5,6,8,9] and toxic heavy metal from the solution [10,11]. Thus this study aims to investigate the adsorption capability of commonly available green algae *U. lactuca* on from solution and effects of initial dyes concentration, contact time, the concentration of algae biomass, and pH.

2. Material and Methods
2.1. Material
2.1.1. Preparation of reagents
Methylene blue C.I.52015 (Merck) is used as a dye source. The dye stock solution was prepared by dissolving accurately weighted dye in the distilled water to the different initial concentration. Dye concentration determined was performed colorimetrically using a Halo UV-VIS Spectrophotometer RB-10. The absorbance of colors was read at 663 nm. The initial pH was adjusted with concentrated Hydrochloric Acid (Merck) or Sodium Hydroxide (Merck).

2.1.2. Preparation of adsorbent
*U. lactuca* (green algae) was collected from Sepanjang Beach, Kemadang, Tanjungsari, Gunungkidul Regency, Special Region of Yogyakarta during November 2017. The collected algae were washed with sea water, tap water, and then distilled water many times. The washed algae were oven dried at 100 °C for 2 h. The dried algae were ground as a powder using an electric mill and sieved to uniform particle size 310 μm.

2.2. Methods
Adsorption experiment was carried out in batch condition. A series of 250 ml Erlenmeyer flask containing 200 ml dye solution of a known initial concentration of 25 mg/L were prepared at room temperature (25 ± 2 °C). Weighted amounts (1.25 g/L) of dry algae biomass were added to each flask and agitated continuously using an orbital shaker. The pH of the mixture was kept without measurement. Equilibrium process is directly correlated with time. The sample was drawn at suitable time intervals 5, 10, 20, 30, 40, 50, 60, 70, 80, 90, 100, 110, and 120 min, and then centrifuged for 10 min at 2000 rpm. The left out the concentration of dye in supernatants were analyzed using the spectrophotometer by observation the absorbance changes at a wavelength of 663 nm.

Effect of pH was tested at pH 5, 6, 7 and 8. To verify the effect adsorbent amount, different weight of algae biomass amounts was variated 0.5; 1; 1.25; 1.5; and 2 g/L. The dye removal from aqueous solution depends on the initial MB concentration that carried at different concentration (15, 20, 25, 30, and 35 mg/L).

Biosorption capacity (qe), the number of dye adsorbed per gram of biosorbent (mg/g) can be calculated as follows:

\[
q_e = \frac{(C_o - C_e) V}{m}
\]
Where $C_0$ is initial dye concentration (mg/L), $C_e$ is the equilibrium concentration of dye (mg/L), $V$ is the volume of solution (L), and $m$ is the mass of biosorbent (g). Percentage of dye removal can also be displayed by the percentage of dye removal as follow:

$$\text{Dye removal (\%)} = 100 \left( \frac{C_0 - C_e}{C_0} \right)$$

3. Results and Discussion

3.1. Effect of contact time

Contact time is the most important issue affecting the efficiency of biosorption. The removal of dye color by *U. lactuca* as monitored at various contact times that shown in figure 1. The adsorption efficiency increases with a rise in contact time up to 110 min, after which it is less constant. Majority of adsorption onto adsorbents was achieved after the first 110 min. The percentage removal of dyes from methylene blue solution increase in contact time and is supported in published studied [2,5,6,9]. As contact time increases, percentage removal also increases initially, but then gradually approaches a constant value. These changes in the rate of removal may be because initial adsorbent sites are vacant and a solute concentration gradient is high [2]. The higher biosorption at the initial contact time may be associated with the driving force of dye into the surface of *U. lactuca* [9].

3.2. Effect of adsorbent dosage

The dye removal values at the variation of *U. lactuca* dosage shown in figure 1. A rise in adsorbent quantity led to a decrease in dye removal and is supported in revealed studied [9]. However, this study is not in agreement with results obtained that supported in publishing studied [5,6] where the sorption capability increased with increasing adsorbent dosage. High biosorbent amounts are known to cause cell agglomeration and subsequent reduction in intercellular distance and produce “screen effect” among a dense layer of cells, resulting to the “protection” of binding sites from metal ions. The obtained knowledge was agreement that supported in publish studied [9,12], the lower biosorbent metal percentage at high adsorbent concentrations [10].

![Figure 1. Effect of contact time and adsorbent amount on the percentage dye removal](image)

3.3. Effect of initial concentration of dye

The effect of dye initial concentration on adsorption of MB was investigated over a spread of pH from 15 to 35 mg/L under constant parameter, pH 7, adsorbent dose 1.25 gr/L, contact time 110 min, and temperature 25 °C ± 2 °C. The solution was agitated continuously using an orbital shaker. The effect of variation of dye initial concentration at dye removal percentages is represented in Figure 2. The result is in accordance with research conducted that supported in publish studied [5], that rate of dye removal
decrease with increase in initial concentration. This is because of the formation of the monolayer at the lower initial concentration of dye over the surface of adsorbent [5].

![Figure 2. Effect of dye initial concentration on the percentage dye removal](image1)

**Figure 2.** Effect of dye initial concentration on the percentage dye removal

### 3.4. Effect of initial pH

The effect of initial pH on adsorption of MB was investigated over a variety of pH scale from 5 to 8 under constant parameter, MB concentration 25 mg/L, adsorbent dose 1.25 gr/L, contact time 120 min, and temperature 25 °C ± 2 °C. The pH was adjusted using 0.1 M HCl and 0.1 M NaOH. The solution was agitated continuously using an orbital shaker. The effect of variation PH at dye removal percentages are represented in Figure 3. These results are in accordance with research conducted that supported in publish studied [5,8,9], where an increasing percentage dye removal gradually increased with pH value. The adsorption of dye on biomass surface is controlled by ionic attraction [8]. This Biosorption alga has principally been attributed to the cell wall properties whatever both electrostatic attraction and complexion will play a task. Algae cell wall often consisting of protein and carbohydrates give a functional group like amino, hydroxyl, carboxyl, sulfate, etc., which can act as binding sites for metals [5,6,13]. At low pH value, a fewer anionic adsorption site on the dried *U. lactuca* was generated, and sorption was unfavorable, probably because excess H⁺ competing with dye molecule for sorption site on dried *U. lactuca*. The surface algae cell all that are liable for binding of dye molecules [5].

![Figure 3. Effect of pH on the adsorption of percentage dye removal](image2)

**Figure 3.** Effect of pH on the adsorption of percentage dye removal
4. Conclusion
The experiment conducted in this study focused on the adsorption dye into *U. lactuca*. The ability of the *U. lactuca* in removing the dye color was dependent on contact time, alga biomass, dye concentration, and pH. The optimum adsorption was found to occur at around pH 8; contact time 110 min; adsorbent dose 1.25 g/L; initial concentration 25 mg/L. The maximum percentage dye removal value was 91.92%. The result showed that green algae *U. lactuca* could be used as biosorbent material for dyes removal from aqueous solution.

5. References
[1] Oladipo M A, Bello I A, Adeoye D O, Abdulsalam K A and Giwa A A 2013 Sorptive Removal of Dyes from Aqueous Solution: A Review *Adv. Environ. Biol. J.* 7 3311–27
[2] El Nemr A, Abdelwahab O, Khaled A and El Sikaily A 2006 Biosorption of Direct Yellow 12 from aqueous solution using green alga *Ulva lactuca* *Chem. Ecol.* 22 253–66
[3] Stefanakis A, Akratos C S and Tsihrintzis V A 2014 Treatment of Special Wastewaters in VFCWs *Vertical Flow Constructed Wetlands* (Elsevier) 145–64
[4] Attia A A, Rashwan W E and Khedr S A 2006 Capacity of activated carbon in the removal of acid dyes subsequent to its thermal treatment *Dyes Pigments* 69 128–36
[5] Mane R and Sabale A Biosorption of Methylene Blue and Malachite Green From Binary Solution onto Ulva lactuca *Int. J. Curr. Microbiol. Appl. Sci.* 3 295–304
[6] Tahir H, Sultan M and Jahanzeb Q 2008 Removal of basic dye methylene blue by using bioabsorbers Ulva lactuca and Sargassum *Afr. J. Biotechnol.* 7 2649–55
[7] Pratiwi D, Indrianingsih A W, Darsi C and Hernawan 2017 Decolorization and Degradation of Batik Dye Effluent using *Ganoderma lucidum* *IOP Conf. Ser. Earth Environ. Sci.* 101 012034
[8] Dahlia M E M 2013 Evaluation of non-viable biomass of Laurencia papillosa for decolorization of dye waste water *Afr. J. Biotechnol.* 12 2215–23
[9] El Sikaily A, Khaled A, Nemr A E and Abdelwahab O 2006 Removal of Methylene Blue from aqueous solution by marine green alga *Ulva lactuca* *Chem. Ecol.* 22 149–57
[10] Ghoneim M M, El-Desoky H S, El-Moselhy K M, Amer A, Abou El-Naga E H, Mohamedein L I and Al-Prol A E 2014 Removal of cadmium from aqueous solution using marine green algae, Ulva lactuca *Egypt. J. Aquat. Res.* 40 235–42
[11] Ibrahim W M, Hassan A F and Azab Y A 2016 Biosorption of toxic heavy metals from aqueous solution by Ulva lactuca activated carbon *Egypt. J. Basic Appl. Sci.* 3 241–9
[12] Esposito A, Pagnanelli F, Lodi A, Solisio C and Vegliò F 2001 Biosorption of heavy metals by *Sphaerotilus natans*: an equilibrium study at different pH and biomass concentrations *Hydrometallurgy* 60 129–41
[13] Srinivasan A and Viraraghavan T 2010 Decolorization of dye wastewaters by biosorbents: A review *J. Environ. Manage.* 91 1915–29