Effective Adsorptive Removal of Methylene Blue from Water Using Wood- Plastic Composite Containing High Density Polyethylene and Wood Particles

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Abstract

In this study, the ability to remove methylene blue cation pigment using wood-plastic composite containing high density polyethylene and wood powder as a recycled material was studied. The effect of some important parameters such as pH, adsorbent amount and contact time were investigated. Adsorption efficiencies for methylene blue was maximized at alkaline pH. Adsorption capacity increased with increasing adsorbent amount and contact time. The value of R^2 in Langmuir model was equal to 1 and the separation factor for 0.5 and 1 g of adsorbent were 0.09 and 0.1, respectively. Given that the methylene blue adsorption data were more consistent with the Langmuir isotherm model, it can be stated that the wood-plastic composite probably has uniform adsorption surfaces and the adsorption process occurred in homogeneous system on the adsorbent surface. Based on the results of this study, it was observed that this composite is a suitable adsorbent for removing methylene blue from aqueous solutions and used as a purifying agent in the decolorization of effluents containing pigments. This adsorbent is recyclable and is cost-effective to remove dye from textile industry wastewater.

Keywords**:** Wood plastic composite, Adsorption, Methylene blue, Langmuir Isotherm

1- Introduction

In the recent years, the development of industry has increased the production of industrial wastewater and sometimes environmental pollution. Dyes are one of the most important pollutants in industrial wastewater, which are generally used in the textile, paper, cosmetics, food, pharmaceutical and leather industries [1]. These industries produce large amounts of industrial waste-water with high water consumption. Due to their complex structure, dye molecules are very resistant to biodegradation and will cause many health problems, including skin allergies, allergies, cancer, mutations, etc. [2]. Colored water is not only aesthetically unpleasant, but also reduces the penetration of light into the water and thus reduces the photosynthetic efficiency of aquatic plants. Methylene blue, also known as Methylthioninium chloride, is a cationic chemical dye that is a dark green, odorless, and solid powder at room temperature that gives a blue solution when dissolved in water. Methylene blue is widely used as a redox oxidation indicator in chemistry. Solutions of this substance turn blue when exposed to the oxidizing medium, but become colorless if exposed to the reducing agent. This cationic pigment is one of the most common dyes used in the textile industry [3]. Due to the characteristics and problems caused by paints, it is necessary to remove artificial colors from wastewater before discharging it to the environment and protect the environment. The different methods to remove dye from wastewater including physical, chemical and biological methods such as coagulation and flocculation, precipitation, adsorption, membrane filtration, electrochemical techniques, ozonation were used in past years [4-6]. Due to the low biodegradability of dyes, conventional biological wastewater treatment processes are not effective in the treatment of colored wastewater, so colored wastewater is usually treated by physical or chemical methods [7]. The adsorption process is one of the physicochemical processes that has been developed over the past years due to its low initial cost, easily operation, flexibility and simplicity for wastewater treatment to remove dyes, organic matter and metals. An ideal adsorbent for wastewater treatment should have features such as environmentally friendly, has a high adsorption capacity and recyclability, and also the adsorbed contaminants should be easily removed from its surface [8]. The adsorbent used in this research is a plastic wood composite that contains 55% by weight of wood particles and 45% by weight of high density polyethylene polymer, which was actually used as a recycled material after the application and the effect of important parameters in adsorption experiments such as contact time, adsorbent amount and pH effect were investigated. Wood-plastic composite is a compound whose main matrix consists of thermoplastic polymers and wood particles, which are generally melted and mixed above the melting temperature of polymers, and used in the manufacture of various wood products such as street protection, floors, facades, billboards, park tables, benches, sidewalks, etc. Wood is a natural absorber that is available in large quantities and at very low prices. Also, polymer adsorbents are considered by different researchers due to their wide surface area, high mechanical stability and ability to remove environmental contaminants in the adsorption process [9]. In recent years, polymers with functional groups of carboxylic acid, amine, hydroxyl and sulfuric acid have been processed, which have been widely used in the removal of dyes, heavy metals and pesticides [10, 11].

2- Experimental

2-1- Materials

High density polyethylene (HDPE), 62N07 with melt flow index (MFI) value of 7 g/10 min (2.16 kg at 190°) was purchased from Lorestan Petrochemical Co., Iran. Commercial Irganox 1010 (powder with molecular weight of 1178 g/mol) and Chinox 168 (powder with molecular weight of 646.94 g/mol) antioxidants were provided from BASF Co., USA and Double Bond Chemical (DBC) Co., Taiwan, respectively. Anti-flame powder (Sb_2O_3) with melting point of 656 °C was supplied from Satrbetter Co., China. Calcium stearate powder (melting point of 150 °C) and paraffin wax (CH36WAX, melting point of 105 °C) were provided from Chimiaran Co., Iran. Maleic anhydride grafted polyethylene 1040 (MAPE) was obtained by ExxonMobil Co., USA, with MFI of 1g/10 min. Waste poplar flour (PF) was supplied from a local sawmill in Tehran, Iran. The sawdust was milled down to particle size of 60 mesh and then dried at 100 °C for 24 h to less than 2% moisture content. Methylene blue (molecular weight of 319.85 g/mol), hydrochloric acid (molecular weight of 6.46 g/mol) and sodium hydroxide (molecular weight of 39.997 g/mol)) were purchased from Merck and double distilled water was used for the experiments.

2-2- Wood plastic composite preparation

Wood-plastic composite prepared and characterized according to our previous work [12]. Appropriate amount of HDPE was mixed with MAPE for 1 minute in a 60 mL internal mixer (Brabender, GmbH & Co., Germany) with the rotor speed of 60 rpm at 180 °C as processing temperature. Then, other additives including antioxidants (2.5 wt%, 2:1 w/w ratio of Chinox 168 to Irganox 1010), anti-flame (4 wt%), calcium stearate (1.5 wt%) and paraffin wax (3 wt%) were added and the mixing continued for another minute. The PF (55 wt%) was added and mixing continued for appropriate time. The resulting wood-plastic composite was used for the removal of methylene blue.

2-3- Drawing of calibration curves

500 ml of 500 ppm methylene blue stock solution was prepared with double distilled water and different concentration (10, 20 and 30, 50, 100 ppm) solution were prepared from stock solution. The resulting solution was analyzed with UV−vis spectrophotometer at 665 nm to obtain the calibration curve.

2-4- Absorption experiment

For the absorption studies different parameters including pH (4, 7 and 9), contact time (10, 30, 60 and 90 minutes) and the amount of adsorbent (0.5 and 1 g) was investigated using a shaker at a speed of 150 rpm at room temperature (Discussion section). After certain shaking times, the solutions were filtered through filter paper and their adsorption was measured using a UV Visible spectrophotometer at 665 nm. Dye adsorption capacity (Q_t, mg/g) and dye removal percentage (% R) by adsorbent at any time were calculated using the following equations, respectively [13]:

$$Q_t = \frac{(C0-Ct)V}{m}$$

$$(\%)\ \text{Removal} = \frac{(C0-Ct)}{C0} \times 100$$

In the above equations C_0 and C_t are the initial concentration and the concentration at equilibrium (mg /L), V is the volume of solution (L), and m is the adsorbent weight (g).

3- Result and Discussion

3-1- Investigation of effective factors on adsorption of methylene blue pigment using wood-plastic composite as adsorbent

3-1-1- pH effect

For the evaluation of methylene blue adsorption experiment at different pH, 25 ml of different concentration of methylene blue including 10, 20 and 30 ppm poured into three separate Erlenmeyer and pH of solutions was adjusted to 4, 7 and 9 with HCl (0.1 N) and NaOH (0.1N) and the adsorption of the resulting solution was analyzed with UV−vis spectrophotometer at 665 nm. Then 1 gr of the wood-plastic composite was added to each Erlenmeyer and shacked at 150 rpm at room temperature for 90 minutes. After the specified time, mixture filtered and adsorption of filtrate was measured by UV−vis spectrophotometer at 665 nm [14]. The result was shown in figure 1.

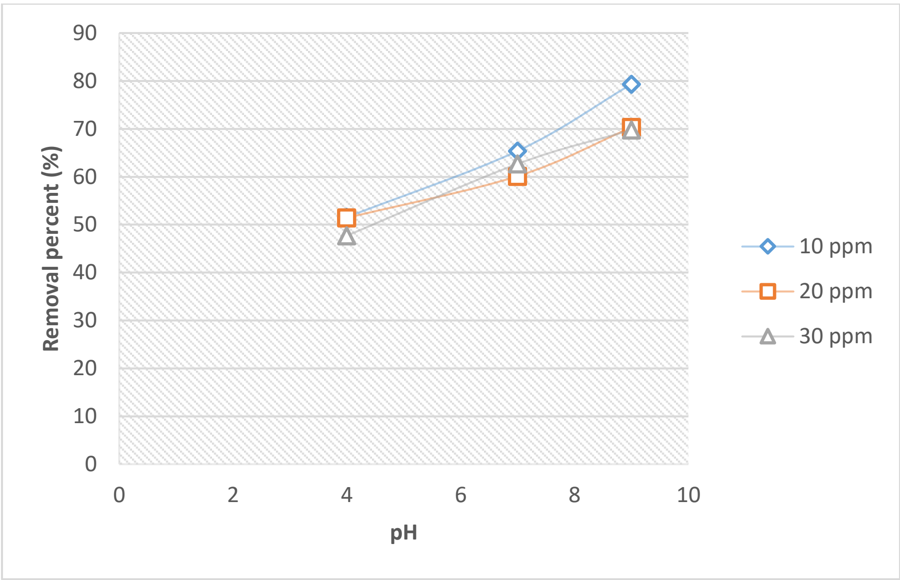

Figure 1. The effect of pH on the amount of methylene blue adsorption

According to Figure 1, with increasing pH, the number of negatively charged sites increases due to the presence of OH ions. As a result, the electrostatic attraction between the adsorbent and the pigment cations and consequently the removal of methylene blue from the aqueous solution increases. So, for studies of subsequent effects, pH = 9 was selected as the optimal pH [15].

3-1-2- Contact time effect

Contact time effect was performed at optimal pH at different times of 10, 30, 60 and 90 minutes. According to Figure 2, at concentrations of 10, 20 and 30 ppm the absorption of methylene blue dye increases with increasing of the time.

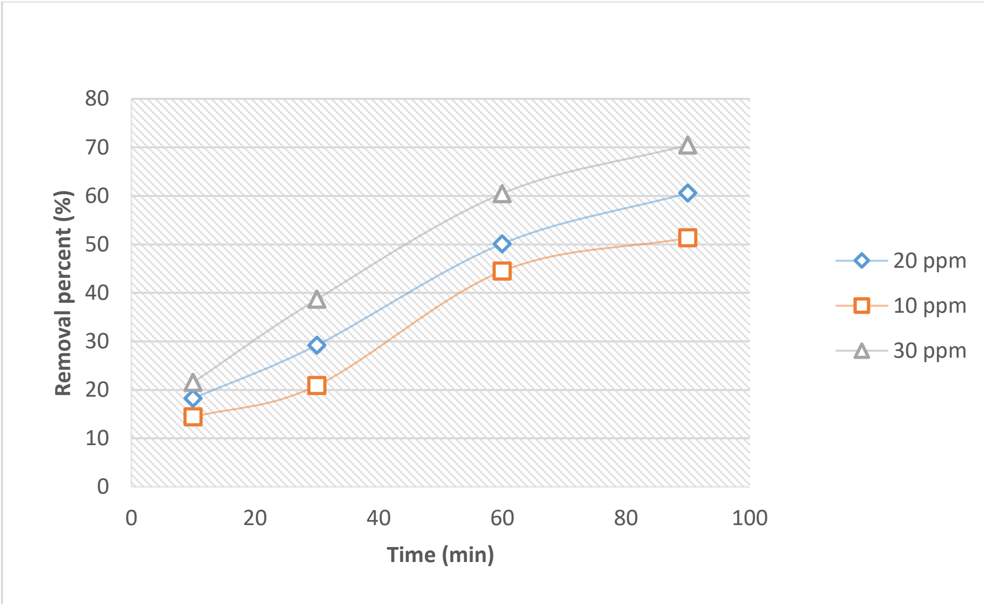

Figure 2. Effect of contact time on the absorption of methylene blue

The adsorption mechanism in a heterogeneous adsorbent is based on surface adsorption and increasing the time increases the contact of the pigment with the adsorbent surface and thus increases its adsorption [16].

3-1-3- Amount of adsorbent effect

To evaluate the amount of adsorbent, 0.5 g and 1 g of adsorbent was added to three different concentrations of methylene blue solution and shacked for 90. According to Figure 3, it can be seen that with increasing the amount of adsorbent, the removal efficiency of methylene blue increases.

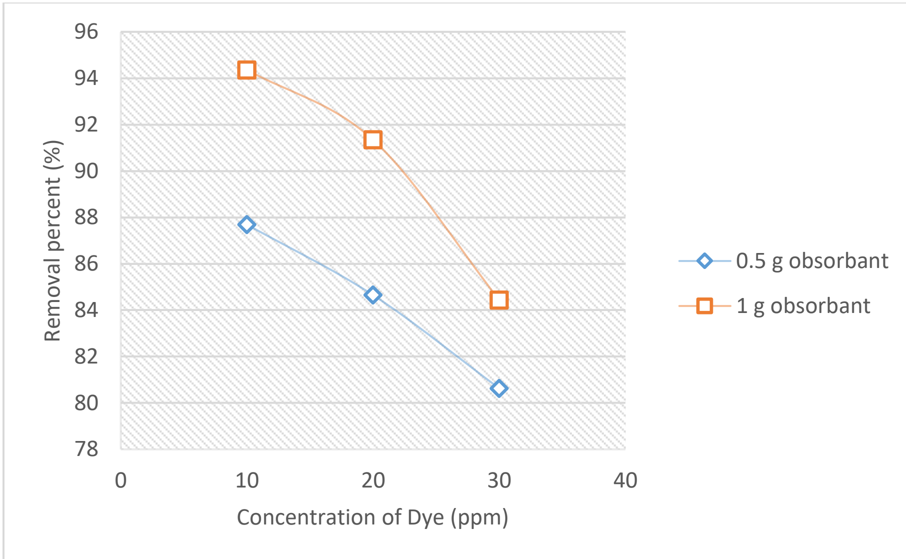

Figure 3. Effect of adsorbent on methylene blue adsorption

This phenomenon can be expressed due to the increase of sites on the adsorbent surface and dye adsorption capacity (mg / g) as the adsorbent mass increases [17].

3-1-4- Investigation of adsorption isotherms

The relationship between the amount of adsorbed substance (Q) and the concentration of that substance in the fluid (C) at temperature T is called the adsorption isotherm at temperature T. Adsorption isotherm has certain constant values that determine the surface characteristics, adsorption dependence and adsorption capacity of different contaminants. Therefore, the adsorption isotherm can provide information about the maximum adsorbents for the adsorption of pollutants, which in turn is useful for designing adsorption processes. In this study, Langmuir and Freundlich isotherm models were used to obtain the adsorption mechanism. The Langmuir model is based on the assumption that adsorption is monolayer without reaction between adsorbed molecules. The Langmuir equation is as follows:

$$\frac{1}{\mathrm{qe}} = \frac{1}{\mathrm{Qmax\ KL}} + \frac{1}{\mathrm{Qmax}}\mathrm{Ce}$$

In the above equation Ce is the equilibrium concentration of dye in solution (mg/L), q_e is the adsorption value (mg/g), Q_{max} is the maximum adsorption capacity (mg /g) and K_L is the Langmuir constant. In Langmuir equation, the dimensionless parameter of separation factor (R_L) is used to describe information from the following equation:

$$R_L = \frac{1}{1+KL\ Ce}$$

The value of R_L determines the type of isotherm, that is reversible ($R_L = 0$), suitable ($0 < R_L < 1$), linear ($R_L = 1$) or unsuitable ($R_L > 1$).

The Freundlich isotherm model is an experimental equation used to describe heterogeneous systems. The Freundlich linear shape is the following equation:

$$\text{Log } q_e = \log K_f + \frac{1}{n} \log C_e$$

In this equation, 1/n indicates the suitability of the adsorption process and the values of K_f and n are obtained from the y-intercept and the slope of the line, which are both Freundlich constants and determine its adsorption capacity and intensity. The magnitude of n is a measure of desirable adsorption. Values of n between 1 and 10 indicate optimal adsorption [14]. The data obtained from the effect of amount of adsorbent were used to calculate the equilibrium parameters such as maximum adsorption capacity and to evaluate the accuracy of each of the Langmuir and Freundlich isotherms. The results are shown in Figures 4 and 5. The calculated data are also given in Table 3-1.

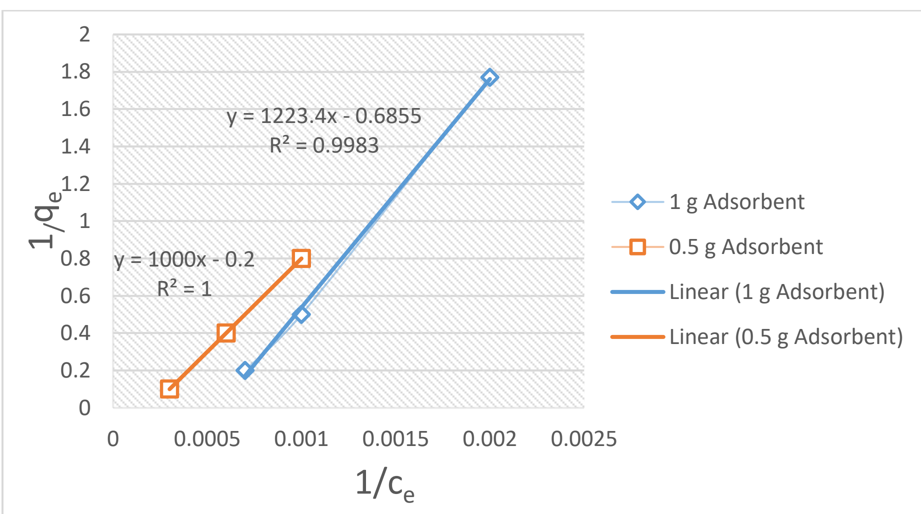

Figure 4. Langmuir isotherm curve

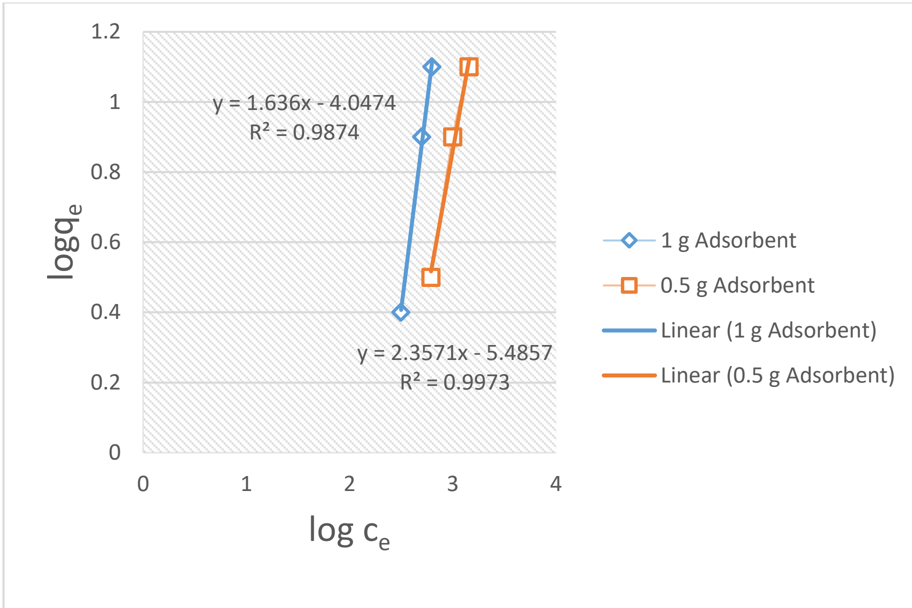

Figure 5. Freundlich isotherm curve

According to Figures 4 and 5, we find that the adsorption data of methylene blue by wood plastic composite are better described by Langmuir isotherm with R^2 value of 1. This fact that the experiment data is more consistent with the Langmuir equation is due to the homogeneous distribution of active sites on the wood-plastic composite surface, because the Langmuir isotherm is based on that the adsorption surfaces are uniform and methylene blue molecules adsorbed on surface of the adsorbent. There is also a good agreement between the obtained results with the Freundlich isotherm model, but the value of R^2 was higher in the Langmuir model. According to the values of separation factor (R_L) 0.09 and 0.1 which was obtained for 0.5 and 1 g of adsorbent indicates the suitability of Langmuir isotherm for the adsorption of methylene blue [17, 18].

Table 1. Langmuir and Freundlich constants

	Langmuir constants				**Freundlich constants**		
Adsorbent	**q_{max} (mg/g)**	**K_L (L/mg)**	**R_L**	**R^2**	**K_F**	**n**	**R^2**
0.5 g	1000	0.2	0.09	1	5.4857	2.3571	0.9973
1 g	1223.4	0.6855	0.1	0.9983	4.0474	1.63	0.9874

Conclusion

Wood-Plastic composite containing high density polyethylene and wood particles as a recycled material as an adsorbent for adsorption of methylene blue cation dye from aqueous samples in the batch system were used. The effect of different parameters including pH, contact time and adsorbent amount were studied. Based on the results, it was found that with increasing pH, the efficiency of dye adsorption increases. By increasing the contact time, the removal efficiency increases. Also, with increasing the amount of adsorbent, the removal efficiency of methylene blue increases. Langmuir and Freundlich isotherm models were used to investigate the adsorption mechanism. According to the results, the methylene blue pigment adsorption mechanism follows the Langmuir monolayer isotherm model and the results of Freundlich isotherm confirm the suitability of the adsorption process. The wood plastic- composite can be used for the removing the pigment from textile industry effluents.

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