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Adsorption of Methylene Blue on spent immobilized biomass of rice straw left after enzyme production

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Abstract

Dyes are widely used in various industries such as the food, pharmaceutical, cosmetic, printing, textile and leather industries. As a result, highly coloured effluents discharged into the environment that affects water transparency. Furthermore, they pose a problem because of their carcinogenicity and toxicity. Therefore, removal of such dyes before discharging them into natural water streams is essential. This research deals with the application of spent immobilized biomass left after enzyme production for the removal of methylene blue from aqueous solutions because of effectiveness of adsorption for dye removal is an alternate to other expensive methods. The effects of four factors namely: pH, dye dosage, adsorbent dosage and temperature on decolorization of methylene blue dye. The results of the analysis show that all selected factors exhibit significant effect on decolorization. Optimum conditions for dye removal were pH 7, temperature 30^{0}C, adsorbent dose 1% and dye concentration of 200 mg/L. By using this adsorbent up to 88% of dye was removed in 48 hrs.

1. Introduction

Dyes are present in wastewater coming from textile dyeing; leather, paper, food and finishing factories (Solozhenkoet al., 1995, Ghawla et al., 2016). Dyes are also used as animal feed preservatives and as disinfectant in aquaculture industry due to their antifungal and antiseptic properties (Kooh et al., 2016). In particular, azo dyes constitute about 50% of dyes normally used in textile industries. They have one or more azo groups (R1-N=N-R2) having aromatic rings mostly substituted by sulfonate groups. These complex aromatic conjugated structures are responsible for their intense color, high water solubility and resistance to degradation under normal conditions (O'neill et al., 2000, Rajaguru et al., 2000, Ghawla et al., 2016). Dye wastewater needs to be disposed properly and should not be discharged directly into water bodies. However such practices are common in some developing countries (Ahmed et al., 2012; Awomeso et al., 2010, Kooh et al., 2016) and these led to severe ecological damages which can spread down to agricultural farmlands or aquaculture industry. This wastewater containing dyes causes aesthetic problems due to the color. The high visibility of dye at low concentration reduces photosynthetic activities of aquatic plant and algae, leading to reduction in dissolved oxygen thereby harming aquatic life (Ahmed et al., 2012, Awomeso et al., 2010). Also these dyes damage the quality of the receiving water because many of dyes released and their breakdown products are toxic, carcinogenic or mutagenic to life forms (Georgiou et al., 2004, Ghawla et al., 2016).

Many technologies have been developed for dye removal from aquatic environments, including physical, chemical, and even biological approaches (Scalese et al., 2016, Robinson et al., 2001, Forgacs et al., 2004). Adsorption is preferred method for treating dye wastewater which is simple and the cost of treatment mainly depends on the choice of adsorbent and the adsorption capacities of the adsorbent. The method can be easily learnt and applied by semi-skilled technician without the need of advanced knowledge, and can be adopted by industries with limited resources (Kooh et al., 2015, Kooh et al., 2016). The choice of adsorbents ranged from abundant plant materials such as invasive weeds e.g. duckweed (Lim et al., 2014) and water fern (Kooh et al., 2015), agricultural residue such as sawdust, rice straw, wheat straw (Hanafiah et al., 2012) and walnut shell (Dahri et al., 2014), as well as biological culture materials such as *Penicilium* (Yang et al., 2011). In present study, fungus immobilized rice straw left after enzyme production (Singh et al., 2011) used as adsorbent for methylene blue dye removal. Effect of pH, dye concentration, adsorbent dose and temperature were studied.

2. Materials and Methods

2.1 Biosorbent preparation

Fungus immobilized microwave alkali pretreated rice straw was used for dye decolorization. *Aspergillus flavus* grown on microwave alkali pretreated rice straw under solid state fermentation for lignocellulolytic enzyme production (Singh et al., 2012) was used as adsorbent. The spent immobilized biomass left after enzyme production dried at 40^0C, dried biomass utilized for dye removal.

2.2. Preparation of dye

Stock solution of methylene blue was prepared by dissolving 1.0 g of each dye in 1000 mL of double distilled water. Working solutions of different concentrations (5–500 mg/L) were prepared by further dilutions. Standard curves were developed through the measurement of absorbance of the dye solutions at 673 nm by UV/Visible Spectrophotometer (T80 UV/VIS Spectrophotometer, PG Instruments Ltd.).

2.3. Batch biosorption studies

Biosorption experiments were conducted in batch mode to investigate the effects of various process parameters such as pH, biosorbent dose, initial dyes concentration, contact time, and temperature on the biosorption of methylene blue. Decolorization of dye was carried out in 150 ml flasks. 50 ml of dye solution was taken in each experiment. Shaking was provided at 150 rpm and 48 hrs studies were carried out for various optimization conditions. Samples were taken after specified time interval and centrifuged at 10,000 rpm in triplicates then analyzed spectrophotometrically at λmax 673nm.

The % biosorption was measured by the following equation:

$$\% \text{ biosorption} = C_o – C_e \times 100 / C_o$$

Where C_o is the initial concentration and C_e is the final concentration. All the experiments were conducted in triplicate.

2.3.1. Effect of pH: In order to study the effect of pH the experiment was setup by varying the pH from 2 to 10 pH of the dye solution, it was adjusted with 0.1 M Sodium Hydroxide (NaOH) and 0.1 M Hydrochloric acid (HCl).

2.3.2. Effect of temperature: In order to study the effect of temperature on decolorization of dye, the experiment was setup by varying the temperature from 20 to 50°C at 5 intervals each.

2.3.3. Effect of Dye dose: In order to study the effect of dye dose on dye removal, experiment was setup by varying dye concentrations i.e. 5ppm, 10ppm, 20ppm, 25ppm, 50ppm, 100ppm, 150ppm, 200ppm, 250ppm, 300ppm, 500ppm and 1000ppm.

2.3.4. Effect of adsorbent dose: In order to study the effect of adsorbent dose on decolorization process experiment was setup by varying adsorbent dose i.e. .5g, 1g, 1.5g, 2g, 2.5g and 5g.

2.4 Adsorption capacity

The amount of dye bound by the biosorbents is known as adsorption capacity and was calculated as follows:

$$Q = v\,(Ci – Cf)\,/m$$

Where Q is the dye uptake (mg of dye per g biosorbent), v is the liquid sample volume (ml), Ci is the initial concentration of the dye in the solution (mg/l), Cf is the final concentration of the dye in the supernatant (mg/l) and m is the amount of the added biosorbent on the dry basis (mg).

3. Results and Discussion

3.1 Effect of pH:

When effect of pH on adsorption of methylene blue was studied, it was observed that the values of adsorption capacity increased with the pH value increasing at the range of 2–4, while, from 4 to 10, the adsorption quantity was approximately constant. Several reasons may be attributed to methylene blue adsorption behavior of the sorbent relative to solution pH. The surface of adsorbent may contain a large number of active sites and the solute (methylene blue ions) uptake can be related to the active sites and also to the chemistry of the solute in the solution. At lower pH values, the surface of adsorbent would also be surrounded by the hydrogen ions which compete with methylene blue ions binding the sites of the sorbent. At higher pH the surface of adsorbent particles may get negatively charged, which enhances the positively charged dye cations through electrostatic forces of attraction. Other adsorbent, such as giant chaff, has the same results about the pH effect on methylene blue adsorption (Waranusantigul et al., 2003, Ghawla et al., 2016). Adsorption of dye increases with pH (Fig. 1) maximum adsorption was took place at 7 pH and then increase at pH 9.

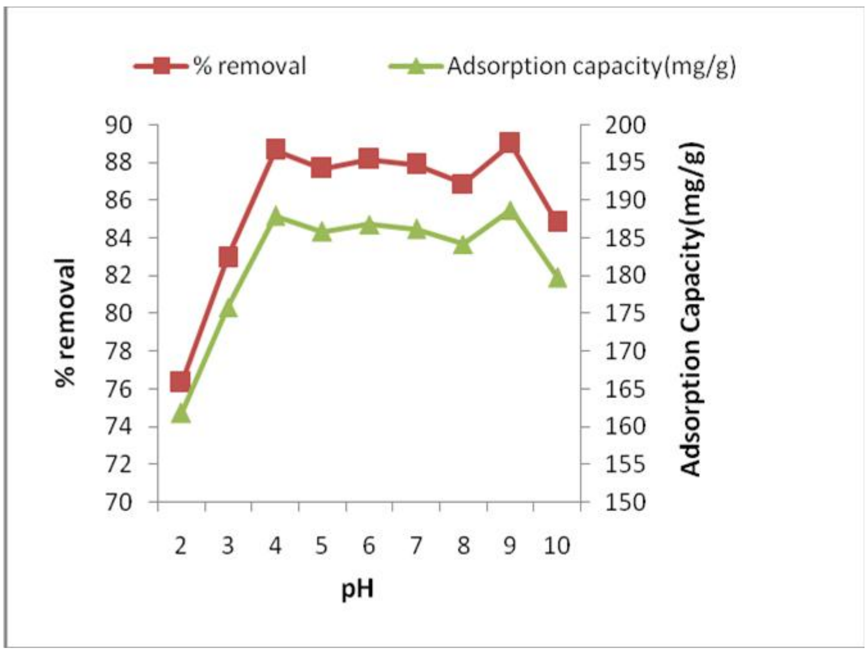

Fig. 1. Effect of pH on % removal and adsorption capacity of methylene blue by *A. flavus* immobilised rice straw

3.2 Effect dye concentration:

The effect of initial dye concentration on adsorption capacity and percent removal is shown in fig. 2. When dye concentration increases up take of dye by adsorbent increased. This is a result of increase in the driving force concentration gradient, as an increase in the initial methylene blue concentrations. In the same conditions, if the concentration of methylene blue in solution was bigger, the active sites of adsorbent were surrounded by much more methylene blue ions; the reaction of adsorption would carry out more sufficiently. So the adsorption quantity for methylene blue increased with the increasing of initial methylene blue concentrations. The percentage of dye adsorption increases initially and attains equilibrium. Other studies have the same results about the initial methylene blue concentration on adsorption capacity and percent removal rate (Otero et al., 2003, Waranusantigul et al., 2003, Vadivelan and Kumar, 2005).

3.3 Effect of adsorbent dose:

The extent of adsorption and removal rate of dye at different biosorbent doses was studied (Fig. 3). It was observed that the value of adsorption capacity decreased with the increase in biomass concentration. The dye's uptake decreased from 55 to 7.49 mg/g for methylene blue due to increase in the biosorbent dose. Maximum biosorption capacity was observed with 0.5 g of biosorbent dose for methylene blue.

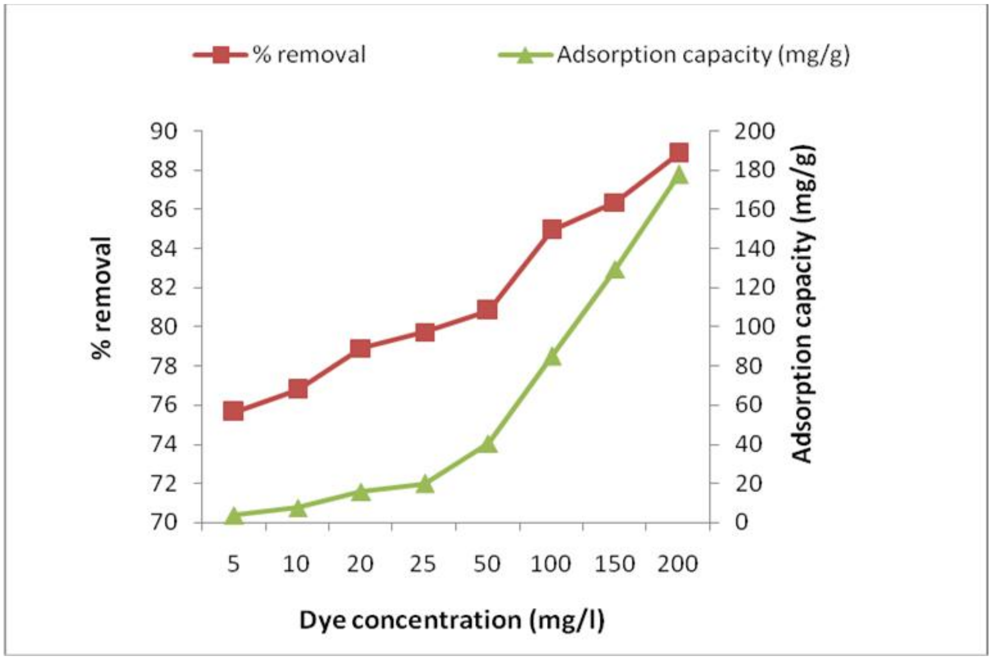

Fig. 2. Effect of dye concentration on % removal and adsorption capacity of methylene blue by *A.flavus* immobilised rice straw

The higher percentage removal at high biosorbent dosage may be attributed to the presence of large surface area of the biosorbent for biosorption. The decrease in biosorption capacity of biosorbent at higher biosorbent dose might be due to aggregation of particles of the biomass which resulted in less binding sites for the dyes. Mohan et al., (2002) reported that adsorbent dose imparted great influence on the biosorption of direct azo dye from aqueous media and maximum dye removal (85%) was observed with 0.5 g dose. A similar effect was previously reported (Waranusantigul et al., 2003, Wang et al., 2005, Vadivelan et al., 2005).

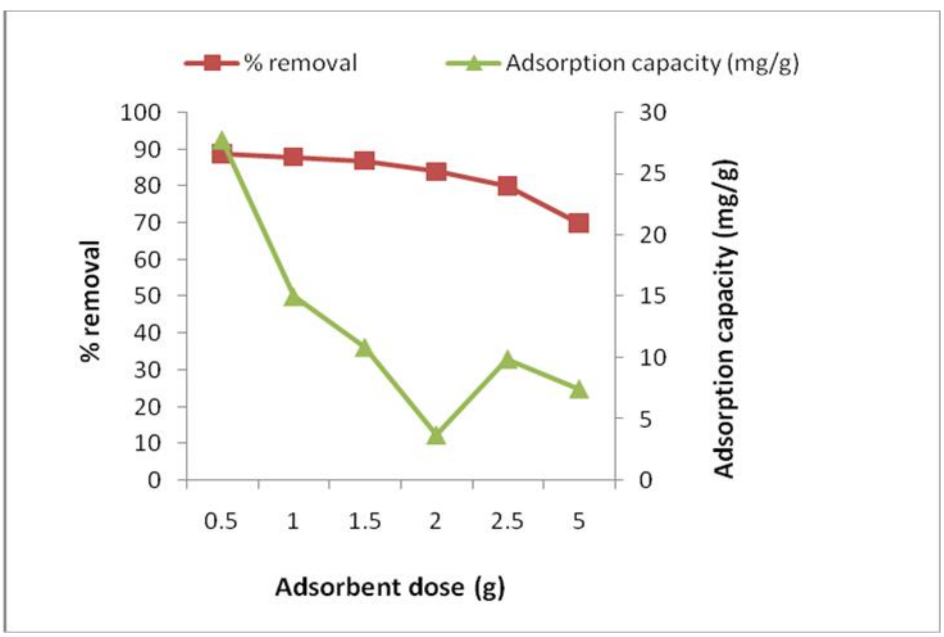

Fig.3. Effect of adsorbent dose on % removal and adsorption capacity of methylene blue by *A.flavus* immobilised rice straw

3.4 Effect of temperature:

Effect of temperature on adsorption: The effect of adsorption rate was carried out at different temperatures from 20°C to 50°C at interval of 5 for adsorbent dosage 0.5g. From the fig. 4, it is shown that removal increases with temperature and then becomes constant. The fact that the percentage dye removal is favored by temperature indicates that the mobility of the dye molecules increases with a rise in the temperature. It can also be said that reaction of dye molecules and surface functional groups is enhanced by increased temperature of reaction Optimum temperature for methylene blue removal was 30°C. This is probably due to a tendency of dye molecules to escape from the solid phase to bulk phase with an increase in temperature of the solution (Gupta et al., 1988) the same trend is observed for all dye concentrations.

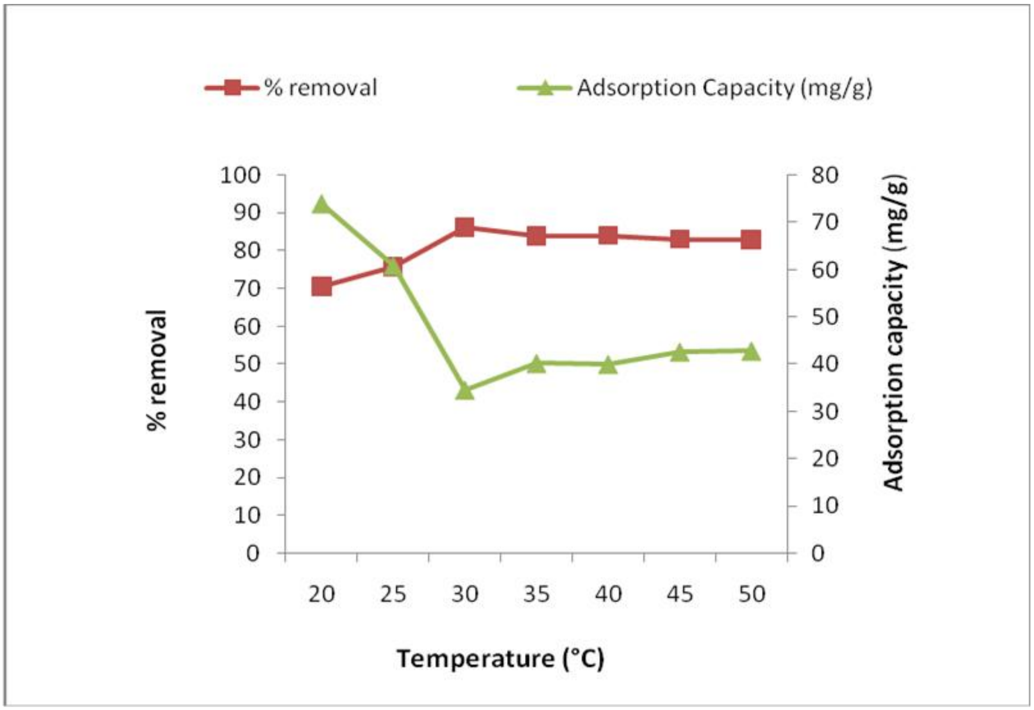

Fig.4. Effect of temperature on % removal and adsorption capacity of methylene blue by *A.flavus* immobilised rice straw.

Conclusion

Adsorption of methylene blue was investigated in batch mode using the spent biomass left after enzyme production. Various parameters were studied such as effect of pH, initial methylene blue concentration, adsorbent dose and temperature were studied. Optimum parameters for dye removal were pH 7, temperature 30 ^{0}C, adsorbent dose 1% and dye concentration of 200 mg/L. Adsorbent used here is a waste of ethanol industry which otherwise require proper waste disposal which could add financial burden on the industry. So it is cheap and effective adsorbent for removal of methylene blue.

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