DEGRADATION OF METHYLENE BLUE IN TEXTILE WASTE WATER USING ACTIVATED SAWDUST AND EGGSHELL BIOSORBENT

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ABSTRACT

Many countries including India are experiencing severe environmental problems due to their rapid industrialization. The textile wastewater contains different dyes which are harmful to the environment. Therefore, wastewater from textile industry has to be treated before being discharged into the environment. The removal of dyes from effluent using adsorption process provides an attractive alternative treatment. The purpose of the present study was to investigate the suitability of using activated sawdust and eggshell as the effective adsorbents used for color removal from the effluent. Batch adsorption studies conducted for the removal of Methylene blue dye in aqueous solution. The removal of dyes at the different contact time, adsorbent dosage, initial dye concentration & pH by activated sawdust and eggshell as adsorbents has been studied and compared. The result shows that both adsorbents are very effective for dye effluent treatment. The maximum adsorption capacity of sawdust was found to be 97.05% and eggshell was 76.47% at neutral pH. The Performance of sawdust adsorbent is good at the pH in between 6 and 7and eggshell adsorbent performed well in the basic pH. The optimum contact time is 30 minutes and optimum adsorbent dosage is 1.5 gram for the sawdust experiment. In the case of the eggshell experiment, the optimum time obtained is 45 minutes and optimum adsorbent dosage is 2 gram. As the initial concentration increases, adsorption efficiency decreases. So, optimum concentration is the lowest concentration of 20mg/l for both experiments. When comparing the both adsorbents, sawdust has high removal efficiency than eggshell at same operating conditions.

Keywords: - *Adsorption, Dye, Textile wastewater, Sawdust, Eggshell, Methylene blue*

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1. INTRODUCTION

Water is most essential unit for all living beings on the earth. Its utility is not limited to human consumption alone, but it is essential for the many productive activities like agriculture, industrial activities, cattle raising, tourism etc. [1]. Limited water resources and increasing urbanization require a more advanced technology to preserve water quality. In industrialized countries, widespread shortage of water is caused due to contamination of ground and surface water by industrial effluents, and agricultural chemicals. Wastewater with high levels of organic and inorganic matter cause several problems when discharged to the environment. It is, therefore, necessary to remove these substances from wastewaters for reducing their harm to environments [2].

Due to rapid industrialization and urbanization a lot of chemicals including dyes, pigments and aromatic molecular structural compounds were extensively used for several industrial applications such as textiles, printing, pharmaceuticals, food, toys, paper, plastic and cosmetics [3]. Textile industry is one of the complicated industries which uses chemicals like dyes, acids, alkalies, starch and surfactants etc., for their processes. Textile industry is a very diverse sector in terms of raw materials, processes, products and equipment and has very complicated industrial chain [4].

In textile industry there are different processes of colour impregnation in fibres, which uses great amount of detergents, dye, microfiber, and inorganic salts. These processes produce huge volume of wastewater containing toxic pollutants leading to the contamination of natural water bodies. Most of the dyes used in the textile industry are monoazo, diazo and triazo dyes, and considering their chemical stability and negative influence on the ecological systems, the regulation of color removal in industrial effluent is a current issue of discussion all over the world [5]. Synthetic dyes are commonly used in most of the textile industries. Over 7 lakh tones of about 10,000 different types of dyes and pigments are produced worldwide [6].

Methylene Blue is the most commonly used material for dying cotton, wood, and silk with molecular weight 373.9 g. It is a heterocyclic aromatic chemical compound with the molecular formula C16H18N3SCl. Methylene Blue is first prepared by Caro in 1876 as an aniline derived dye form textiles. At room temperature it appears as a solid, odorless, dark green powder. So that it yields a blue solution when dissolved in water.

Currently there are various treatment processes such as oxidation process, coagulation, filtration, ozonation, membrane filtration, hydrogen peroxide, reverse osmosis processes are employed in determining the removal of color from the wastewater. These processes become ineffective due to the major disadvantages like high cost, sludge generation and handling difficulties of waste generated. The general bio-treatment processes like activated sludge process are also relatively ineffective due to the low biodegradability of the dyes. Due to these problems in the treatment methods, further investigations have been fed up. The adsorption process provides effective and alternative effect in removal of Color and other parameters in treatment of wastewater using low cost adsorbents which are readily available and inexpensive[7].

Adsorption is a phase transfer process that is widely used in practice to remove substances from fluid phases (gases or liquids). The solid material that provides the surface for adsorption is referred to as adsorbent; the species that will be adsorbed are named adsorbate. In water treatment, adsorption has been proved as an efficient removal process for a multiplicity of solutes. Here, molecules or ions are removed from the aqueous solution by adsorption onto solid surfaces. By changing the properties of the liquid phase (e.g. concentration, pH) adsorbed species can be released from the surface and transferred back into the liquid phase. This reverse process is referred to as desorption.

There is a large variety of adsorbent materials have been tested to absorb the dye molecules and then remove the color from wastewater included natural or synthetic adsorbents. Biosorbents is one of the most favourable choices in recent research for the application of removing toxic pollutants. The advantages of using biosorbents is to remove toxic pollutants in high efficiency, produce minimum chemical or biological sludge, ability to regenerate biosorbents, and the possibility of metal recovery following adsorption.

In recent years there are numerous researches focused on developing cheaper materials with more effective adsorption to remove dyes from wastes such as, fibres, banana pitch , biogas residual slurry , chitosan, hard wood , rice husk , silica, bentonite, peat, orange peel, jackfruit seed, eggshell, sawdust etc. Sawdust has proven to be a promising effective material for the removal of dyes from wastewaters. If sawdust could be used as adsorbent, both the environment protection and wooden industry could benefit. Moreover, it is actually an efficient adsorbent that is effective to remove many types of pollutants, such as dyes, oil, salts, heavy metals. Sawdust is largely composed of cellulose pectin; hemi-cellulose, lignin, etc. Sawdust is alternative as an adsorbent for its abundance in nature, non-toxicity and bio-degradability. The removal of dyes from wastewater using adsorption process by sawdust provides an alternative treatment [8].

Many researches have been done to make use of egg shell in different applications such as, fertilizers, feed additive and adsorption heavy metals and organic compounds from waste water [9]. The chemical composition of egg shell (94% calcium carbonate, 1% magnesium carbonate, 1% calcium phosphate and 4% organic material), as well as the porous nature of egg shell structure (7000-17000 pores) makes it an attractive material to serve as an adsorbent agent. Furthermore, the inner eggshell membrane has a good adsorption characteristics due to its composition (polysaccharides fibers and collagen like protein), which containing substituting group sites such as hydroxyl, amine and sulfonic groups can react with dye [10].

This study deals with the degradation of Methylene Blue in textile wastewater using activated sawdust and activated eggshell as adsorbent materials. The objectives of the study are to find out the optimum values of pH, contact time, adsorbent dose and initial concentration and also to compare adsorption capacity of both the adsorbents.

2. METHODOLOGY

2.1 Preparation of activated sawdust

The collected saw dust sample is cleaned with water and dried in the atmosphere. After drying it is soaked with the concentrated sulfuric acid for 24 hours. Then washed with distilled water and again soaked in the sodium carbonate solution for 30 minutes to remove residue acid .Then washed with distilled water and oven dried for one hour at 100°c.Sieved the sample through 0.3mm sieve. The obtained sawdust then treated with H_2O_2 for 30 min. And finally add 10% con. HNO_3 with constant stirring for 40 min. Washed and oven dried at 100°c for another 24 hours.

2.2 Preparation of activated eggshell

The collected eggshell washed with tape water to remove surface adsorption then sun dried. The grinded eggshell then socked with $H_2SO_4$4 solution for overnight to increase adsorption efficiency. Then washed with distilled water till it attained neutral pH and treated with 5% sodium bi carbonate solution for 24 hours in order to remove excess of acid present. Then washed with distilled water to remove dirt and dried at oven at 105°C for 24 hours. Finally allow to pass through 0.3mm sieve.

2.3 Preparation of Methylene Blue solution

Stock solution of Methylene Blue concentration (1000 mg/L) was prepared by diluting 1 gm of Methylene Blue powder in a 1000 ml of distilled water. Later it was diluted to get the test solutions of varying concentrations 5 to 100 mg/L.

2.4 Methylene Blue standard curve

The concentration of methylene blue in the standard solution before adsorption was determined using a double beam UV spectrophotometer at 660 nm. It was found that the standard solutions (5 to 100mg/l) give some absorbance value and which is important for preparing standard curve.

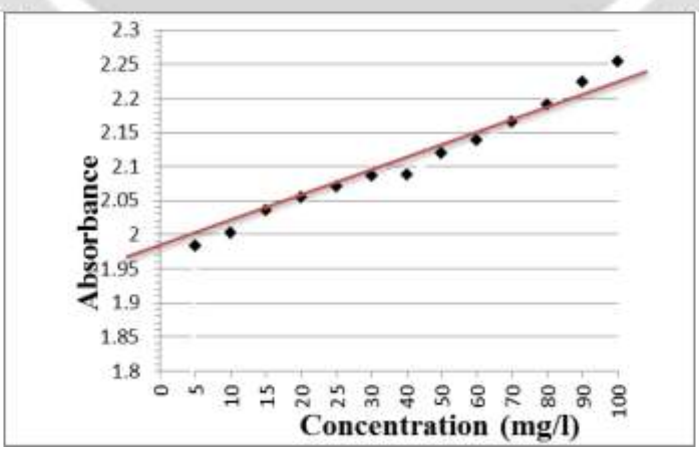

Chart -1: Methylene blue standard curve.

From the Figure 1 we see that, absorbance of standard solution is determined and plotted in the Y axis and concentration of standard solution is plotted in X axis. Value of absorbance and concentration gives a straight line equation which has a slope m, and the value of m is important for determining unknown concentration and absorbance.

2.5 Effect of contact time

To study the effect of contact time 0.5 gm of adsorbent is taken 30 ml of aqueous solution of initial methylene blue concentration 20 mg/L, at known pH 7 and the constant shaking was provided for 40 minutes. Collected the sample at different time intervals like 5, 10, 15, 20, 25, 30, 35, 40, 45, 50, 55 and 60 minutes and filtered. To determine the optimum contact time analyzed the amount of adsorption of each sample.

2.6 Effect of initial concentration

To study the effect of methylene blue concentration 0.5 grams of adsorbent is added to 30 ml of methylene blue solution of concentration 20 mg/L and is keep shaking for optimum time. Procedure is repeated with the 30 ml solution of pH 7 at the different initial concentrations 20, 40, 60 and 80 mg/L keeping the other conditions constant. Then filtered and analyzed for optimum amount of methylene blue concentration.

2.7 Effect of adsorbent dosage

The effect of adsorbent dosage on the amount of methylene blue adsorbed was obtained by agitating 30 ml of methylene blue solution of 20mg/L separately with 0.5, 1, 1.5, 2, and 2.5 grams of adsorbent for optimum shaking time by keeping the other conditions constant. Determined the absorbance of each sample and hence find out optimum dosage.

2.8 Effect of pH on dye adsorption

To determine the effect of pH in the dye removal mechanism, take methylene blue solution of concentration 80 mg/L , add acid in order to reduce pH value 4, 6, and add base to increase the pH to 8, 10 and 12. Freshly prepared hydrochloric acid and sodium hydroxide can be used to change the pH. After setting the pH of the ranges 4, 6, 8, 10 and 12, 50 ml sample solution pipette out in to each flask and add 0.5 grams of saw dust to it and allowed to undergo shaking for optimum time. Determined the absorbance of each sample and hence find out the optimum pH.

3. RESULTS AND DISCUSSION

Activated sawdust and activated eggshell powder used as an adsorbent in the present study to remove methylene blue dye from textile waste water. The results of this study are discussed in the following section.

3.1 Effect of contact time

The study had shown that for sawdust the percentage of dye removal was high. The figure 2 and figure 3 represents the graph of variation of adsorption percentage with contact time of sawdust and eggshell respectively. The data obtained from the adsorption of Methylene Blue on to sawdust, showed that a constant time of 30 minutes was sufficient to achieve equilibrium and adsorption did not change significantly with further increase of time. In the case of eggshell, the maximum adsorption occurred after 45 min and there was almost no adsorption beyond this time because the equilibrium is attained. This was due to the fact that, at the initial stage the number of free adsorption sites was higher, and the slow or constant adsorption rate in the later stage was due to slower diffusion of solute into the interior of the adsorbent.

Table-1: Optimum contact time

Time (minutes)	% Removal (Sawdust adsorbent)	% Removal (Eggshell adsorbent)
5	22.05	16.17
10	47.05	22.05
15	58.82	26.47
20	72.06	30.88
25	77.94	38.23
30	85.29	41.17
35	85.76	47.05
40	85.76	57.35
45	-	61.76
50	-	61.76
55	-	60.29
60	-	60.29

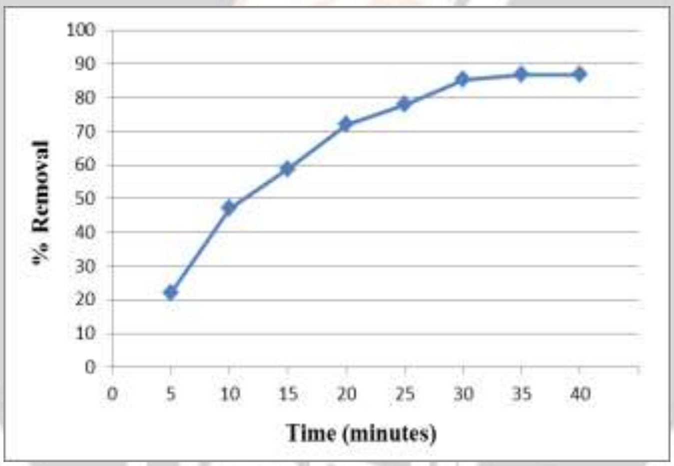

Chart -2: Effect of contact time on sawdust

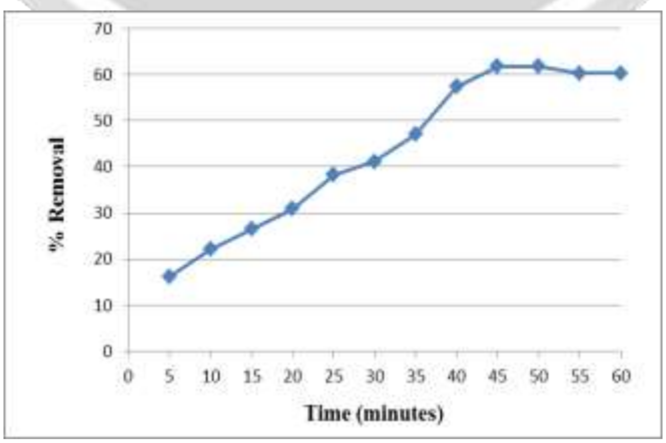

Chart -3: Effect of contact time on eggshell

3.2 Effect of Initial Concentration

From the table 2 it is clear that increase in the dye concentration had caused the decrease in the percentage of dye removal. This is due to lack of sufficient surface area of adsorbent to accommodate much more adsorbate available in the solution. It is observed that sawdust adsorbent degraded 85.29 % of methylene blue at initial concentration of 20 mg/l and in the case of eggshell adsorbent dye removal was 61.76% at same condition. The figure 4 and figure 5 represents the graphical variation of percentage removal with initial concentration of sawdust and eggshell respectively.

Table -2: Effect of initial concentration

Initial concentration (mg/l)	% Removal (Sawdust adsorbent)	% Removal (Eggshell adsorbent)
20	85.29	61.76
40	76.47	48.52
60	72.54	35.29
80	63.97	22.05

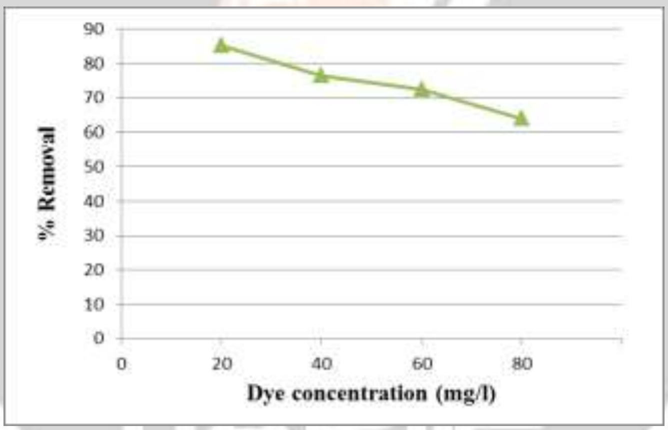

Chart -4: Effect of initial concentration on sawdust

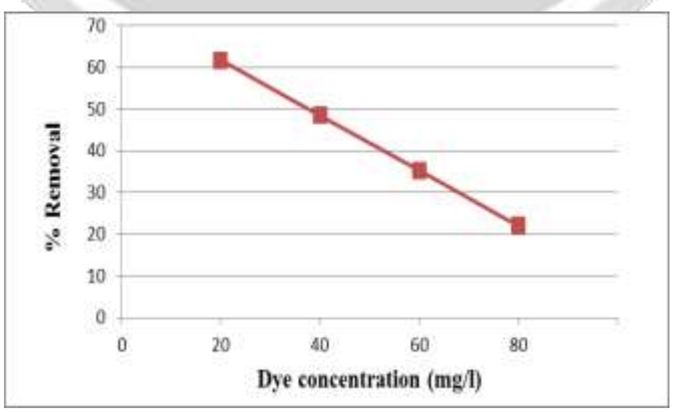

Chart -5: Effect of Initial Concentration on eggshell

3.3 Effect of Adsorbent Dosage

The effect of adsorbent dosage on the adsorption of methylene blue is shown in Table 3. It was observed that adsorption of dye increased with the increase of the amount of adsorbents and then attained a value at equilibrium. The variation of removal percentage with adsorbent dose is clear from the figure 6 and figure 7. The increase of dosage increases adsorbent sites thus surface area of contact with the dyes increases consequently leads to a better adsorption. In the sawdust experiment, 97.05% removal was observed at optimum adsorbent dosage of 1.5g and eggshell adsorbent removed 76.47% of color at optimum dosage of 2g.

Table -3: Effect of adsorbent dosage

Adsorbent Dosage (g)	% Removal (Sawdust adsorbent)	% Removal (Eggshell adsorbent)
0.5	85.29	61.76
1	92.64	66.17
1.5	97.05	70.58
2	97.05	76.47
2.5	-	76.47

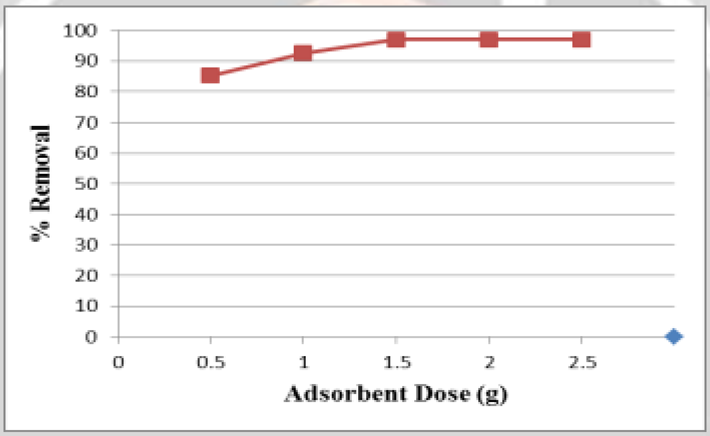

Chart -6: Effect of sawdust adsorbent dosage

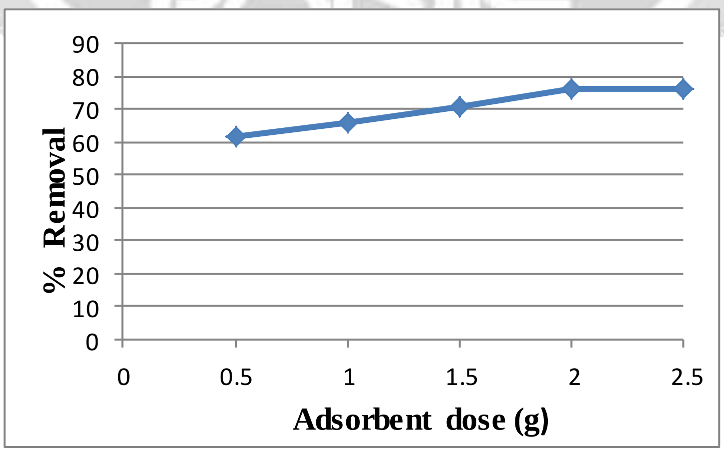

Chart -7: Effect of eggshell adsorbent dosage

3.4 Effect of pH on dye adsorption

The effect of pH on the percentage of the Methylene Blue removal is shown in Table 4 under various other fixed operating conditions. The initial pH of Adsorption medium is one of the most important parameters affecting the adsorption process. In the figure 8 and figure 9 it is seen that the percentage of dye removal was high at the pH of 6 to 7 in the case of sawdust adsorbent and removal is high at pH 10 in the case of eggshell adsorbent.

Table -4: Effect of pH

pH	% Removal (Sawdust adsorbent)	% Removal (Eggshell adsorbent)
4	57.72	19.85
6	64.33	21.69
7	63.97	22.05
8	59.55	36.76
10	50.36	54.41
12	-	45.95

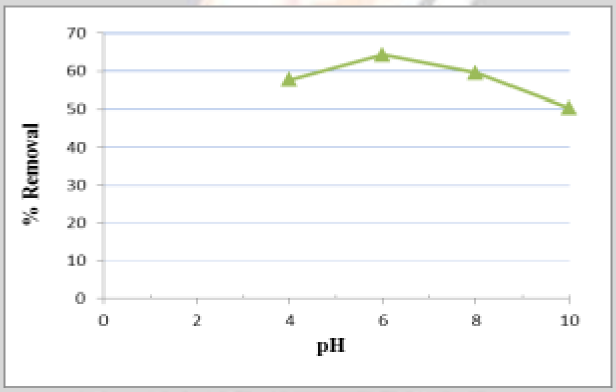

Chart -8: Effect of pH on sawdust

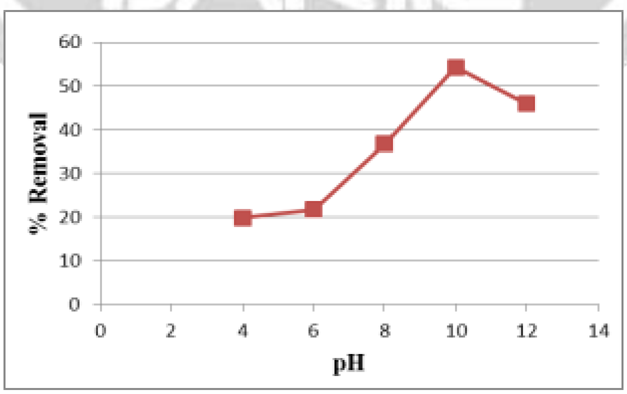

Chart -9: Effect of pH on eggshell

4. CONCLUSIONS

This study entailed the Degradation of methylene blue in textile waste water using activated sawdust and eggshell biosorbent. The results showed that sawdust proved to be a very effective biosorbent in the removal of methylene blue dye from textile waste waters than the eggshell for the same experimental conditions.
It can be also concluded that;

- The biosorption performance is strongly affected by parameters such as contact time, initial dye concentration, adsorbent dosage and pH
- The maximum removal of methylene blue observed was 97.05 % for 1.5g of sawdust adsorbent at neutral solution.
- In neutral solution maximum removal obtained is 76.47% for 2g of eggshell adsorbent
- The optimum conditions of sawdust experiment are 30 minutes of contact time, 1.5 g adsorbent dose and pH of solution in between 6 and 7
- The optimum conditions of eggshell experiment are 45 minutes of contact time, 2 g of adsorbent and pH of the solution is 10.
- It was observed that percentage adsorption of methylene blue decreases with increase in the initial concentration of aqueous solution and the amount of adsorbate adsorbed increases with the increasing of adsorbent dose.

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