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RESEARCH ARTICLE

EFFECTIVE REMOVAL OF TEXTILE DYES USING PRE-TREATED NATURAL ADSORBENTS

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ABSTRACT

Water contamination due to textile industrial discharge is one of major problems of modern world. Azo dyes are major culprits of water pollution due to presence of complicated azo bond. Synthetic wastewater with high concentration of azo dyes has been treated using different adsorbents in this study. This exploration technique is proved to be conservative, appealing and compelling for the removal of dyes from wastewater. In this investigation, pre-treated sugarcane bagasse and chicken eggshell were utilized as an adsorbent for adsorption of azo dyes from wastewater. All the experiments were allowed to run for 160 minutes. These adsorbents satisfactorily served the purpose of application. Eggshells and sugarcane bagasse and presented 79% and 73% removal of reactive red-1 respectively. Removal of direct blue-1 was observed as 96% and 95% using eggshells and sugarcane bagasse respectively.

KEYWORDS

Removal, Adsorption, Sugarcane bagasse, Eggshell, dyes, wastewater.

1. Introduction

Emissions of azo dyes from industrial activities are a major cause of degradation of water quality. Within this context, the developing countries contribute the largest amount of textile wastewater (World Bank Group, 2012). Consistent releases of modern wastewater containing azo dyes in water bodies cause different unsafe consequences for conditional and general well-being. Water is very important to the human body. Every one of your cells, organs and tissues use water to help with temperature regulation, keeping hydrated and maintaining bodily functions (Adeleye et al., 2016). In addition, water acts as a lubricant and cushions your joints. Weighty dyes can bio-accumulate into the earth through natural way of life (Nagarajappa et al., 2017). However, there is an elevated obstruction in this way of getting water due to water pollution. Contamination of Water disturbs the quality of almost every source e.g., surface water (ocean, rivers and lakes) and underground water becoming a widespread issue. Natural phenomenon may also contribute towards pollution of water sources, but human activities contribute largely towards contamination of water bodies (Lofrano and Meriç, 2019).

Moreover, about 10% of dyes used in the textile industry are lost during the dyeing process, and 2% are directly discharged as aqueous effluents into the environment (Essawy et al., 2008). Without adequate treatment, these compounds retain their color and structural integrity under exposure to sunlight, soil, bacteria and sweat, and exhibit a Sulphur cotton and rayon aromatic substrate vetted with sodium sulphide and re-oxidized to insoluble Sulphur containing products on fibre indeterminate structures Vat cotton, rayon, and wool water-insoluble dyes solubilized by reducing with sodium hydrogen sulfite, then exhausted on fiber and re-oxidized anthraquinone (including polycyclic quinones) and indigoids 9 high resistance to microbial degradation in wastewater treatment systems (Hameed and El-Khaiary, 2008). They remain in the environment for longer periods, if discharged without adequate treatment (Soloman et al., 2009; Baban and Ciliz 2010; Robinson et al., 2001).

Ability of azo dyes to deplete dissolved oxygen, inhibitory effect on photosynthesis in water and toxicity for human beings, flora and fauna and transformation along food 2chains are the prime concerns with the dyes effluents. Carcinogenic and toxic aromatic amines are produced if azo dyes are treated anaerobically. Out of total dyes used 60% are azo dyes (Ali et al., 2018; Siddique et al., 2017). Industrial sectors can pollute the surroundings via many ways. A huge extent of dangerous effluent is typically generated in way of means of commercial activity. Dye stuff manufacturing, dyeing and fabric Industries discharged wastewater containing a whole lot of dyes into water bodies (Katheresen et al., 2018). The presence of dyes in water will lessen the mild Penetration, Precluding the photosynthesis of aqueous flora (Paven et al., 2007). Some dyes can cause or reason health trouble including allergy, dermatitis, pores and skin inflammation and most cancers to human and being mutagenic (Paven et al., 2007). Azo dyes are very complex compounds with diversified structures. Azo dyes are very stable biologically and chemically so are resistant to degradation, resistant against microorganisms and water and soap also adds to their stability (Konicki et al., 2017).

Incomplete degradation of microorganism produce poisonous amines if dyes are damaged or break down in sediment (Hamdaoui, 2006). Besides, formation of poisonous cancer agents is a huge trouble while dyes weighted down wastewater is immediately discharged or disposes into our environment and the municipal wastewater vegetation and surroundings (Hamdaoui, 2006). Partial and complete removal or decolorization of toxic dyes can be achieved by biological, physical and chemical processes (Khattar and Shailza, 2009). Biological methods are used in connection with activated sludge processes and membrane bioreactors. Chemical processes include the advanced oxidation of dyes

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with ozone, H_2O_2 which can be run concomitantly under UV-irradiation. These three methods of dyes removal have some limitations and are not economically feasible, but adsorption has advantage over the other methods because of simple design and can involve low investment in term of both initial cost and land requirement (Lai et al., 2014; Wiesmann et al., 2007).

Hence, a most cost-efficient manner of dyes elimination is extra most desirable where adsorption technique comes out to be a powerful and appealing remedy. Adsorption technique is straightforward to operate with easy design, land required, and no results of poisonous materials and advanced elimination of natural waste materials (Lee, 2009). There is huge potential for utilization of the agro-based waste materials e.g., activated carbon, rice husk, sugarcane bagasse, and peanut shells etc. as adsorbents to remove dyes from textile wastewater. After practical work sugarcane bagasse and chicken eggshell both adsorbents are proved very efficient in removing azo dyes from wastewater (Lofrano, 2012). Based on all above discussion this research focused on exertion of sugarcane bagasse and eggshell for removal of azo dyes through adsorption technique because adsorption is feasible and viable option among all other methods of dye removal.

2. Materials and Methods

2.1 Collection and Preparation of Adsorbent

The agro-based adsorbent sugarcane bagasse was acquired locally near Nazimabad an area of city Faisalabad. The sugarcane bagasse was pre-treated by cutting into small pieces and boiled for 3 hours to remove dirt and organic materials, after the boiling process, SCB was dried in an oven at 100 °C for 48 h. After that, the SCB was milled and sieved to obtain a particle size 0.6 mm (Nouren et al., 2017). The eggshell used in the experiment were collected free of charge from different hotels located in Faisalabad, using plastic bags and washed with tape water to remove dirt then dried at 105 °C for 1 h in a convection oven and grounded. Then washed with distilled water till it attained neutral PH to remove dirt and boiled to remove color and dried at oven at 105 ^{o}C for 1 hour and activated in muffle furnace at 7000 ^{o}C for 2 hours. Finally allowed to pass through 0.6 mm sieves.

2.1.1 Preparation of Dye Solution

Two dyes were being studied. For each dye, 1000 mg/L of standard dye solutions were prepared as stock solution and subsequently diluted when necessary. All prepared dye solutions were kept in dark to prevent light degradation.

2.2 Experimental Setup

Experiments were conducted to optimize different parameters for dye removal. The efficiency of prepared solution was measured by conducting experimental study on two Azo dyes "Direct blue" 1 and "Reactive Red" 1. pH and contact time were optimized after conducting experiments on pH range 3-11 and adsorbents dose in the range 0.2 g/100mL and contact time 5,10,20,40,80,160 min. The supernatant was analyzed for its dye concentration using UV-Visible Spectrophotometer.

2.2.1 Pollutants Studied

Two azo dyes employed in this study are very complicated in terms of structures that are difficult to degrade. Direct blue-1 has molecular weight of 992.80 g/mol, λ_{max} of 619 and molecular formula of $C_{34}H_{24}N_6Na_4O_{16}S_4$. Molecular structure of direct blue-1. The molecular weight of reactive red-1 is 717.38 g /mol and molecular formula is $C_{19}H_9C_{l2}N_6Na_3O_{10}S_3$, having maximum absorbance in spectrophotometric analysis at λ_{max}=551. Figure displays the structural formula of reactive red-1.

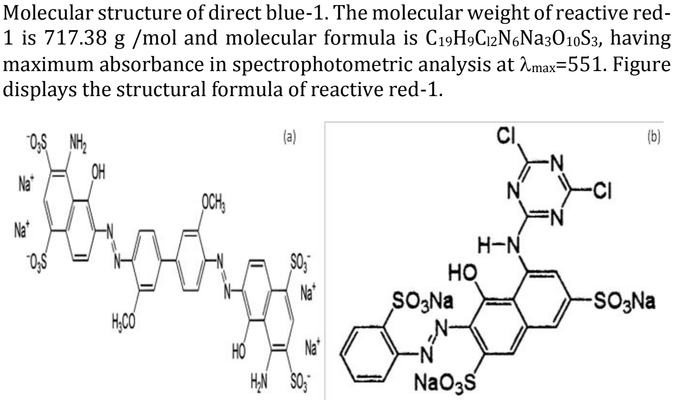

Figure 1: Structural formula of (a) Direct Blue-1 (b) Reactive Red-1

2.3 Response Surface Methodology (RSM)

Response Surface Methodology (RSM) is useful for the modeling and analysis of programs in which a response of interest is influenced by several variables and the objective is to optimize this response (Yeap et al., 2014). The main idea of RSM is to find an optimal response by a sequence of designed experiments. So, RSM was used to optimize the effect of three parameters (pH, contact time and adsorbent dose). These parameters were designated as independent variables, while the percentage removal of reactive red-1 and direct blue-1 was designated as the dependent response variable in the central composite design (CSD). Equation (1) representing the general equation of cubic model

$$Y= \beta_o+ \beta_1A+ \beta_2B + \beta_{12}AB + \beta_{11}A^2 + \beta_{22}B^2+ \beta_{12}AB + \beta_{112} A^2 B + \beta_{122} AB^2 + \beta_{111}A^3 + \beta_{222}B^3 \quad (1)$$

Where,

Y = Response (Percentage Removal)

β_o= intercept

A= 1st independent variable (pH)

B= 2nd Independent variable (Contact Time)

And β_1, β_2, β_3, β_{12}, β_{11}, β_{22}, β_{12}, β_{112}, β_{122}, β_{111}, β_{222} are regression coefficients.

3. Results and Discussions

The experiment was conducted on 50 ppm solutions of direct blue-1 and reactive red-1 dye by using adsorbents sugarcane bagasse and chicken eggshell. It was found that maximum and early time decolorization for direct blue-1 and reactive red-1 occurs at pH=5 and pH=7 respectively. The adsorption rate was rapid during the first 30 min and then continued at a slower rate from 35 to 80 min, and almost reached at level after approximately 160 min of the experiment (Zaghbani et al., 2008). This was due to the fact that, at the initial stage the number of free adsorption sites was higher, and the slow adsorption rate in the later stage was due to slower diffusion of solute. The maximum adsorption occurred after 100 min and there was almost no adsorption beyond this time.

3.1 Reaction Kinetics of Experimental Results

Reaction kinetics experiment was conducted on dye solutions of different concentrations (50ppm, 100ppm, 150ppm). The experiment was allowed to run for 160 minutes in total. Figure 2(a) and 2(b) shows the decolorization behavior of direct blue-1 dye by applied adsorbent sugarcane bagasse and chicken eggshell respectively, and figure 3(a) and 3(b) shows the decolorization behavior of reactive red-1 dye by applied adsorbent sugarcane bagasse and chicken eggshell respectively. It was observed by adding or enhancing adsorbent quantity decolorization occurs more quickly as well as gives better results in terms of percentage removal.

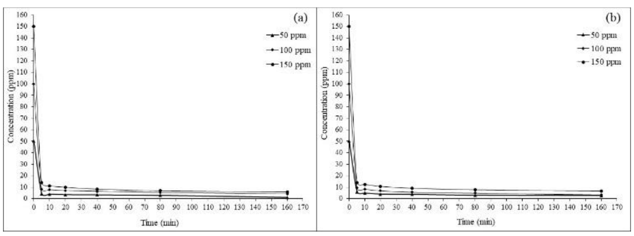

Figure 2: (a) Reaction kinetics of Direct Blue-1 with eggshell (b) Reaction kinetics of Direct Blue-1 with sugarcane bagasse

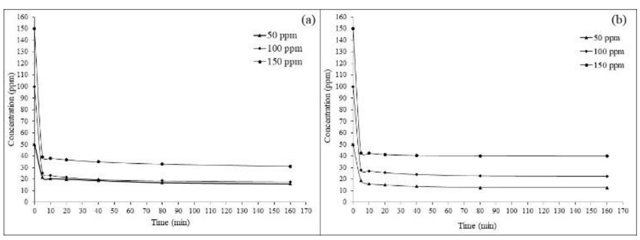

Figure 3: (a) Reaction kinetics of Reactive Red-1 with eggshell (b) Reaction kinetics of Reactive Red-1 with sugarcane bagasse

3.2 Optimization of the process data using RSM

For optimization of relevant processes RSM is being widely used as it explores the relationship between independent variables and dependent variables. The response of dependent variable as a function of independent variables (pH, Contact time and dose) were investigated by central composite design (CCD) using RSM analysis for both the dyes.

3.2.1 RSM analysis of direct blue-1 using chicken eggshell as adsorbent

The final equation in terms of coded factors for direct blue-1 using sugarcane bagasse and eggshell is shown as equation 2. Figure 4 shows that all the data is deviated normally from mean value. There is no significant deviation from mean value.

$$\text{Percentage Removal} = +93.71 - 4.50\times A - 4.22\times B + 0.28\times A\times B - 8.12\times A^2 + 3.92\times B^2 + 2.83\times A^2\times B - 4.77\times A\times B^2 + 8.93\times A^3 + 5.40\times B^3 \quad (2)$$

Contour map and 3D surface shows the pattern of dependent variable (Percentage removal) as a reaction to free factor (Contact time and pH). 3D surface shows the pattern in 3D view and Contour map shows that pattern in type of forms. The figure 5 (a) shows that most extreme percentage removal that was achieved at 30 minutes and also toward the finish of investigation (120 min) at pH of 5 for direct blue-1 to 7 for reactive red-1. Figure 5 (b) shows 3D surface that depicts the rate of percentage removal increases as contact time increases.

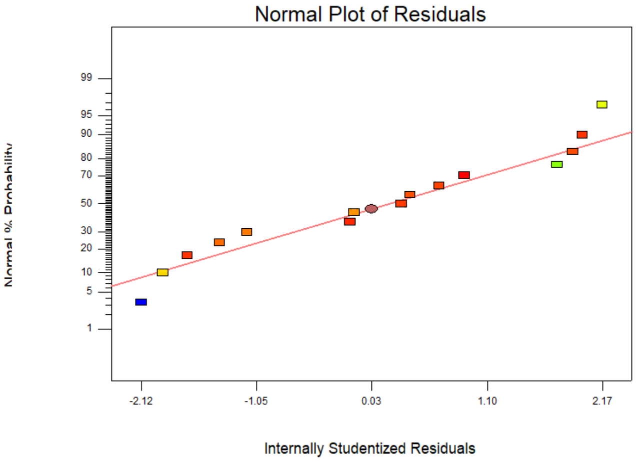

Figure 4: Normal Plot of data for direct blue-1 with eggshell

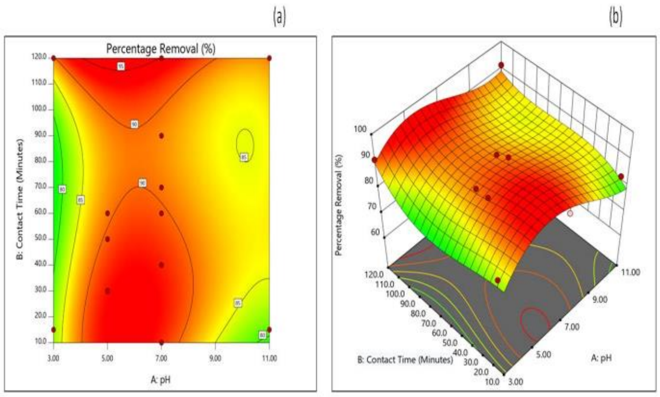

Figure 5: (a) Contour map and (b) 3D surface of removal (%) as a function of pH and contact time (hours) for direct blue-1 dye with eggshell

3.2.2 RSM analysis of direct blue-1 using sugarcane bagasse as adsorbent

Equation (3) represents the final equation in terms of coded factors for direct blue-1 using sugarcane bagasse and eggshell.

$$\text{Percentage Removal} = +93.65 - 18.41\times A + 15.16\times B - 4.85\times A\times B - 16.65\times A^2 - 6.38\times B^2 + 1.32\times A^2\times B + 8.18\times A\times B^2 + 26.81\times A^3 - 7.31\times B^3 \quad (3)$$

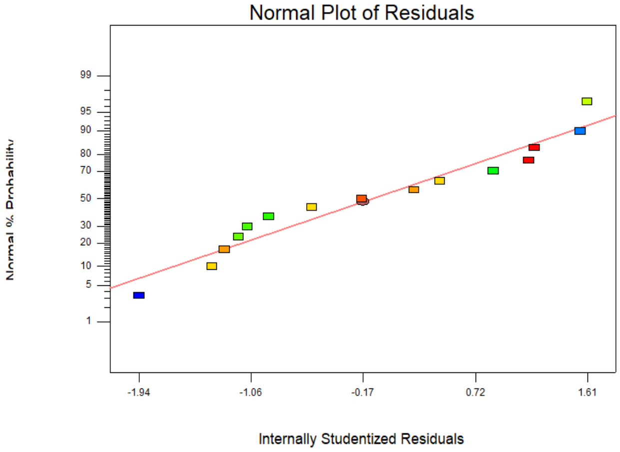

Figure 6: Normal Plot of data for direct blue-1 with sugarcane bagasse

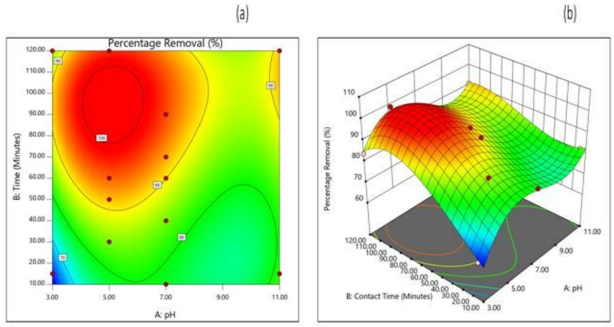

Figure 7: (a) Contour map and (b) 3D surface of removal (%) as a function of pH and contact time (hours) for direct blue-1 dye with sugarcane bagasse

3.2.3 RSM analysis of reactive red-1 using eggshell as adsorbent

The normal probability plots of the residuals and the plots of the residuals versus the predicted response for surface r and internally studentized residuals are shown in figure 8 respectively. A check on the plots in figure 10 revealed that the residuals generally fall on a straight line implying that the errors are distributed normally. Equation (4) represents the final equation in terms of coded factors for direct blue-1 using sugarcane bagasse and egg-shell.

$$\text{Percentage Removal} = +87.02 - 22.60\times A + 2.65\times B - 3.19\times A\times B - 17.81\times A^2 + 1.89\times B^2 - 4.04\times A^2\times + 15.18\times A\times B^2 + 13.65\times A^3 + 8.25\times B^3 \quad (4)$$

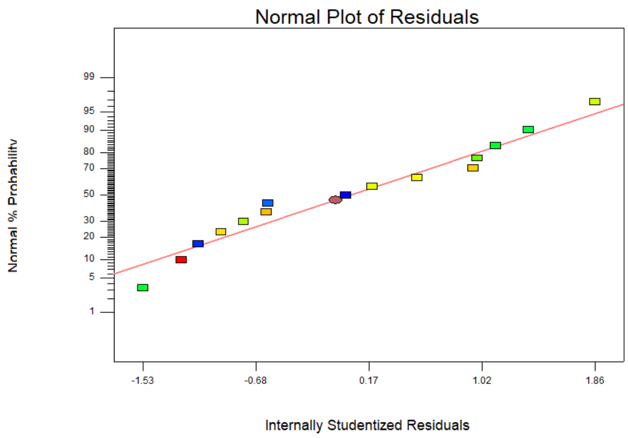

Figure 8: Normal Plot of data for reactive red-1 with eggshell

Figure 9 (b) response surface plot (3-D plot) is the graphical illustration of the potential relationship between three variables. It is obtained by plotting two independent variables on the x- and y-scales, and the response (z) variable is represented by a smooth surface (3-D surface). Similar figure 9 (b) contour plots and 3-D surface plots are useful for establishing the response values but in a more concise manner.

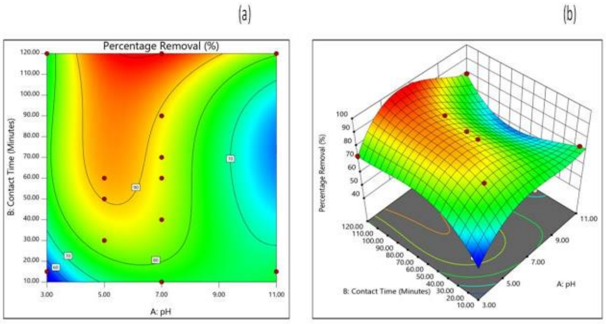

Figure 9: (a) Contour map and (b) 3D surface of removal (%) as a function of pH and contact time (hours) for reactive red-1 dye with eggshell

3.2.4 RSM analysis of reactive red-1using sugarcane bagasse as adsorbent

Equation (5) represents the final equation in terms of coded factors for direct blue-1 using sugarcane bagasse and egg-shell.

$$\text{Percentage Removal} = +91.16 - 6.2\times A + 24.62\times B - 3.82\times A\times B - 10.96\times A^2 - 5.95\times B^2 + 3.41\times A^2\times B + 7.19\times A\times B^2 + 3.04\times A^3 - 17.97\times B^3 \quad (5)$$

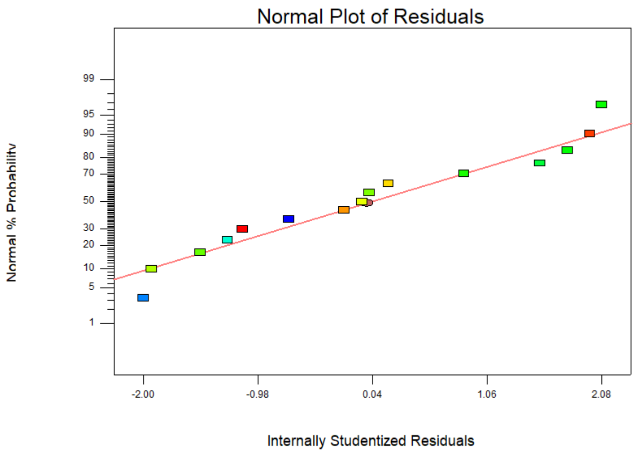

Figure 10: Normal Plot of data for reactive red-1 with sugarcane bagasse

The figure 11 (a) shows that maximum percentage removal was achieved at 110 minutes and also toward the finish of investigation (120 min) at pH of 7 for reactive red-1. Figure 11 (b) shows 3D response surface that exhibits the rate of percentage removal increases with the increase in contact time.

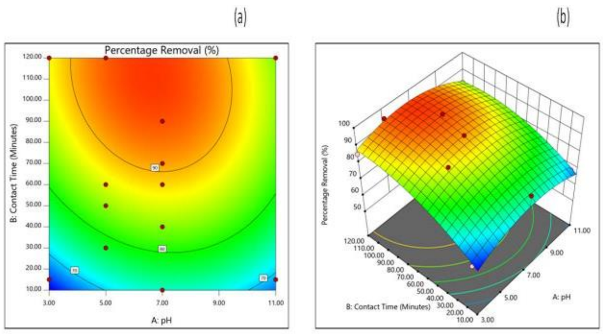

Figure 11: (a) Contour map and (b) 3D surface of removal (%) as a function of pH and contact time (hours) for reactive red-1 dye with sugarcane bagasse

This indicated that both models were valid as the lack-of-fit were not significant. Low C.V. values for binary direct blue-1 (3.91%) and reactive red-1 (1.67%) showed the good precision and reliability of the experiments. Adequate signal to noise ratio were also observed for both models, showing that both models can be used to navigate the design space. For binary dye solution, all the coefficients were significant. The 3D surface plot for binary dye solution of interaction between pH and initial dye concentration at contact time of 120 min and 0.2 g/L of sorbent dosage. The maximum percentage uptake of dye was observed when initial dye concentration was at minimum point while pH was at maximum point within the studied range. As the initial dye concentration increased the percentage uptake decreased. Decrease in percentage uptake at high initial dye concentration might be due to insufficient of available binding sites. Maximum percentage uptake was observed at pH 5-7 was due to the increased of negatively charged sites as the pH of the dye solution increased. According to Hameed et al. (2009), the surface charge may be positively charged at lower pH.

4. CONCLUSION

The optimum pH for the removal of the direct blue-1 dye was observed 5. While on the other hand optimum pH for the removal of the reactive red-1 dye was observed 7. For direct blue-1 dye the maximum removal was observed after 120 min and for reactive red-1 the maximum removal observed after 90 min. Sugarcane bagasse is found to be more efficient in removing direct blue-1 dye as compared to eggshell. On the other hand eggshell is more efficient then sugarcane bagasse for removal of reactive red-1 dye.

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