

Removal of reactive dyes from aqueous solutions by a non-conventional and low cost agricultural waste: adsorption on ash of Aloe Vera plant

Mohammad Malakootian¹, Hossein Jafari Mansoorian*², Ahmadreza Yari³

1) Research Center and Environmental Health Department, School of Public Health, Kerman University of Medical Sciences, Kerman Iran.

2) Health Promotion Research Centre, Zahedan University of Medical Sciences, Zahedan, Iran

3) Research center for environmental pollutants, Qom university of medical sciences, Qom, Iran

*Author for Correspondence: h.mansoorian@yahoo.com

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ABSTRACT

Dyes are an important class of pollutants and disposal of them in precious water resources must be avoided. Among various methods adsorption occupies a prominent place in dye removal. The aim of this study is to evaluate adsorption of dye Reactive Red 198 and Blue 19 (RR-198 & RB-19) on to Aloe Vera plant ash from aqueous solutions.

In this research Aloe Vera ash was prepared at laboratory conditions and then after shredding, screened by ASTM standard sieve with 60 -200 mesh sizes and the effects of pH (3-12), adsorbent dose (0.1-1 g/L), contact time (10-60 min), initial dye concentration (10-160 mg/L) and temperature were investigated in the experiment. In different samples Dye concentration was measured by spectrophotometer at 592 nm and 520 nm wavelength for RR198 and RB19 respectively. Also the Langmuir and Freundlich adsorption isotherms were determined in order to describe the relations between the colored solutions and the adsorbent.

The results of this study showed that acidic conditions were more conducive to enhance the hydrolysis rate than basic ones as the decomposition was optimum at pH 3. The adsorption rate of RR-198 and RB-19 dyes was increased by increasing of initial dye concentration, increasing of adsorbent dose in 0.1 to 0.4 mg/L. Dye solution was decolorized in a relatively short time (20 min). The efficiencies for RR-198 and RB-19 reactive dyes were 82.68% and 90.42% respectively. The maximum adsorption capacity (q_{max}) has been found to be 80.152 mg/g for RR-198 reactive dye and 88.452 mg/g for Blue 19 reactive dye. Adsorption isotherms were examined by Freundlich and Langmuir isotherm that finally showed the Freundlich multilayer isotherm has better accordance with data.

The results indicate that Aloe Vera ash plant as a natural and inexpensive adsorbent is a suitable adsorbent for the adsorption of textile dyes.

Keywords: Agricultural waste, Aloe Vera leaves ash, Dye RR-198 & RB-19, Adsorption Isotherms

INTRODUCTION

One of the important classes of the pollutants is dyes that are considered to be particularly dangerous organic compounds for the environment [1]. Once the dyes enter the water it is no longer good and sometimes difficult to treat as the dyes have a synthetic origin and a complex molecular structure which makes them more stable and difficult to be biodegraded. Dye molecules comprise of two key components: the chromophores, responsible for producing the colour, and the auxochromes, which can not only supplement the chromophore but also render the molecule soluble in water and give enhanced

affinity (to attach) toward the fibers [2, 3]. Dyes may be classified on the basis of their solubility: soluble dyes which include acid, mordant, metal complex, direct, basic and reactive dyes; and insoluble dyes including azoic, sulfur, vat and disperse dyes. It is worthwhile noting that the azo dyes are the one most widely used and accounts 65–70% of the total dyes produced [4]. Nearly 40,000 dyes and pigments are listed which consist of over 7000 different chemical structures. Most of them are completely resistant to biodegradation processes. Overall at present there are more than 100,000 commercial dyes with a rough estimated production of 7×10^5 – 1×10^6 tons per year [5].

Recent studies indicate that approximately 12% of synthetic dyes are lost during manufacturing and processing operations. Approximately 20% of these lost dyes enter the industrial wastewaters [6]. Dyes from dyeing processing wastewaters are found in the wastewater streams of industrial processes, including paint manufacture, dyeing, textiles, paper and pulp, tannery and others [7]. The textile dyeing factories are the major sources of environmental pollution due to the discharge of wastes to water resources. Normally, colour is discernible at a dye concentration higher than 1 ppm and the average concentration of 300 ppm has been reported in the effluents from the textile production [5]. Reactive dyes are one of the most common used synthetic dyes in the dyeing industry. These types of dyes are highly soluble in water and have reactive groups which can form covalent bonds between dye and fibre [2, 6]. Reactive dyes are those most commonly used to dye cotton and other cellulosic fibres, wool and nylon [8]. These dyes form a covalent bond with the fiber and contain chromophoric groups such as azo, anthraquinone, triarylmethane, phthalocyanine, formazan, oxazine, etc. Their chemical structures are simpler, absorption spectra show narrower absorption bands, and the dyeing are brighter, making them advantageous over direct dyes [9]. Large amounts of these dyes are discharged in the water courses, due to the hydrolysis, low fixation rate, etc. in developing countries that can have bad effects on water and human bodies [10]. Thus removing color from wastewater is more important than treating other colourless organics and there is an urgent need for textile wastewater to develop effective methods of treatment because small amounts of dye are clearly visible and detrimental to the water environment [11, 12]. Generally, the methodologies to treat dye wastewater are classified in four categories: (i) physical (ii) chemical (iii) biological and acoustical, radiation, and electrical processes [3, 10]. Although there are many methods for the removal of the dyes, it is difficult to treat the wastewater by using traditional methods, because most of the synthetic dyes are stable to light, chemicals and biological treatment, etc. [13]. Among these methods, adsorption can be thought to be the most efficient process for the treatment of wastewater due to its low-cost and ease of operation as well as greater efficiency and the main adsorbent used in dye removal is activated carbon

[14]. An activated carbon was developed from coconut shell fibers, characterized and used for the removal of methylene blue (basic) and methylorange (acidic) dyes from wastewater successfully. Activated carbons were prepared from the agricultural solid wastes, silk, cotton hull, coconut tree sawdust, sago waste, maize cob and banana pith and used to eliminate heavy metals and dyes from aqueous solution [15, 16].

Although activated carbon seems to be an efficient adsorbent, it has high operating costs because activated carbons are expensive due to their regeneration and reactivation procedures [14]. For this reason, many studies revealed that other alternative adsorbents. In recent years, growing research interest in the use of cheap and eco-friendly adsorbents such as agricultural residues as an alternative substitution of activated carbon for the treatment of contaminated wastewater containing different classes of dyes [5, 12]. The by-products from the forestry and agricultural industries could be assumed to be low-cost adsorbents since they are abundant in nature, inexpensive, require little processing and are effective materials. These materials are available in large quantities and may have potential as sorbents due to their physico-chemical characteristics and low-cost. Agricultural residues are lignocellulosic substances which contain three main structural components: hemicelluloses, cellulose and lignin [17, 18].

Aloe Vera is an abundant by-product of the agricultural industry that is either used in the production of a latex substance and drugs. Aloe Vera contains amino acids minerals, vitamins, enzymes, proteins, polysaccharides, biological stimulators, antiseptic agents and anti-inflammatory fatty acids. Mature plants can grow as tall as 2 and a half inches to 4 feet with the average being round 28 to 36 inches in length. The Aloe Vera plant is grown in warm tropical areas. Internationally, Aloe Vera can be found in the United States, Mexico, India, South America, Central America, the Caribbean, Australia, Africa and Iran [19, 20]. In this work, the adsorption capacity of Aloe Vera plant ash for the removal of Reactive Red 198 and Blue 19 (RR-198 and RB-19), a synthetic dye, were examined. The effects of pH, adsorbent dosage, contact time and initial dye concentration, for the adsorption were investigated. The Langmuir isotherm model in the studied

concentration range of RR-198 and RB-19 at different temperature was also tested.

MATERIALS AND METHODS

Reagent and Materials

The dye used in the experiments was RR-198 and RB-19, which was provided from Alvan Sabet Company of Iran and the other chemicals used in these experiments were the product of the Merck Company (Darmstadt, Germany). Double distilled water (DDW) was used throughout the study. Table 1 shows the structure of the investigated dyes. These dyes are azo and contain anionic sulphonate groups. Stock solutions of dyes were prepared by dissolving the powder in double distilled water. Dye solutions of different initial concentrations were prepared by diluting the stock solution in appropriate proportions.

Preparation of the Adsorbent

Initially, Aloe Vera leaves gel extracted and the leaves washed with distilled water for several times to remove all the dirt particles. Then the adsorbent (Aloe Vera leaves) was dried in an oven at 105 °C for 10 h, then maintained at 300°C for 2 h to preparation of ash. The prepared ash were sieved (with a stainless steel strainer (149-74µm, Mesh 100-200) and stored in a bottle.

Dye Removal Experiments

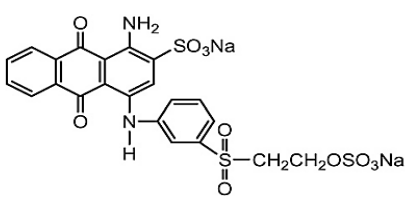
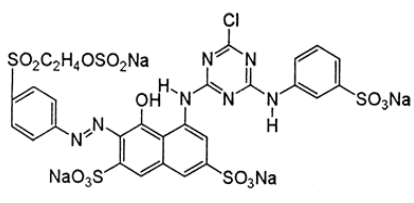
The batch mode of operation was carried out in order to measure the adsorption rate. Wastewater samples containing dye in different concentrations

(10-160 mg/L) will be prepared as synthetic in the laboratory. In the first stage with constant dye concentration, contact time and adsorbent dose, pH optimum is determined among the pHs of 3-12. The pH of the dye solutions was adjusted to optimum value using 0.1 M HCl and/or NaOH. In the next stage with constant dye concentration, contact time and pH, the optimum adsorbent is determined among the amounts of 0.1–1g/L and particle size (149–74µm) on the adsorbent were studied for a contact time of 60 min. In all experiments the samples will be mixed for 80 min at the speed of 100 rpm by orbital shaker. Taken samples are smoothed by the filter 0.45 mm until concentration of dye in the treated effluents was determined by using UV Spectrophotometer (IRMECO of Model U2020) at wavelength of the maximum absorbance of each dye that indicated in Table 1. The dye removal efficiency was determined using the following expression:

$$R(\%) = \frac{C_o - C_t}{C_o} \times 100 \quad (1)$$

Where C_o and C_t represent the initial and final (after adsorption) concentrations of dye in mg/L, respectively.

Table 1: Characteristics of the used dyes

Characteristics	Reactive Blue 19 (RB-19)	Reactive Red 198 (RR-198)
Chemical Structure		
Molecular Formula	$C_{22}H_{16}O_{11}N_2S_3Na_2$	$C_{27}H_{18}ClN_7Na_4O_{15}S_5$
Molecular Weight	626.5 g/mol	967.5g/mol
λ_{max}	592 nm	520nm

RESULTS AND DISCUSSION

Effect of pH

pH plays a major role in the adsorptive removal of dyes from aqueous solution and is an important factor controlling the surface charge of the adsorbent and the degree of ionization of the materials in the solution [21]. Dyes exist in the ionic form in the aqueous system. As charged species, the degree of their adsorption onto the biosorbent surface is primarily influenced by the surface charge on the biosorbent which in turn, is affected by the solution pH. The pH of the solution influences the properties of biomass materials, affects the adsorption mechanisms and dissociation of the dye molecules [22]. The results showed that the maximum biosorption of both dye RR-198 and RB-19 was observed at low pH 3 (Fig.1). At lower pH, the biosorbent surface turned out to be positively charged and electrostatic attraction develops between positively charged biomass and negatively charged anionic dyes. However, at basic pH, adsorption decreases due to the presence of hydroxyl ions which show competition with dye anions for binding sites [23]. Also at $pH > pH_{pzc}$ (pH positively charged=3), the decreased dye adsorption capacity of Aloe Vera leaves ash was attributed to the repulsive interaction between the positively charged Aloe Vera leave ash and negatively charged (sulphonate groups) dye molecules. pH_{pzc} is the pH at which the net surface charge on adsorbent is zero. The adsorbent surface has a net positive charge at $pH < pH_{pzc}$, while having a net negative charge at $pH > pH_{pzc}$ [13, 19]. Ardejani *et al.* [24] examined the effect of initial pH on adsorption of Direct Red 80 from aqueous solution onto almond shells. As pH increased from 3 to 12, the adsorption capacity decreased from 20.5 to 18.8 mg/g. The maximum uptake of dye was observed at pH 3.0. Hence, pH 3.0 is selected in the subsequent experiments.

Effects of Adsorbent Dosage

The effect of different adsorbent dosage 0.1–1 g per 1000 ml solution under identical operating conditions for the removal of the dye is shown in Fig. 2. It was observed that at higher adsorbent dose, removal increases sharply within 30 minutes of adsorption. 83.46% for RR-198 and 92.25% for RB-19 removal is observed within initial 30 min of contact with 0.4 g of ash of Aloe Vera leave. An increase of adsorbent dosage from 0.1 to 0.4 g/L causes an increase adsorption rate by 23 % and 25% for RR-198 and RB-19 respectively. This is due to the fact that, increase in adsorbent dosage increase area available for adsorption. At lower adsorbent doses, increase in removal is proportional to the increase in adsorbent dose. At a

higher dose increase in removal of RR-198 and RB-19 is less than proportional dose due to continuous depletion of reactive dye molecules in aqueous phase [23, 25].

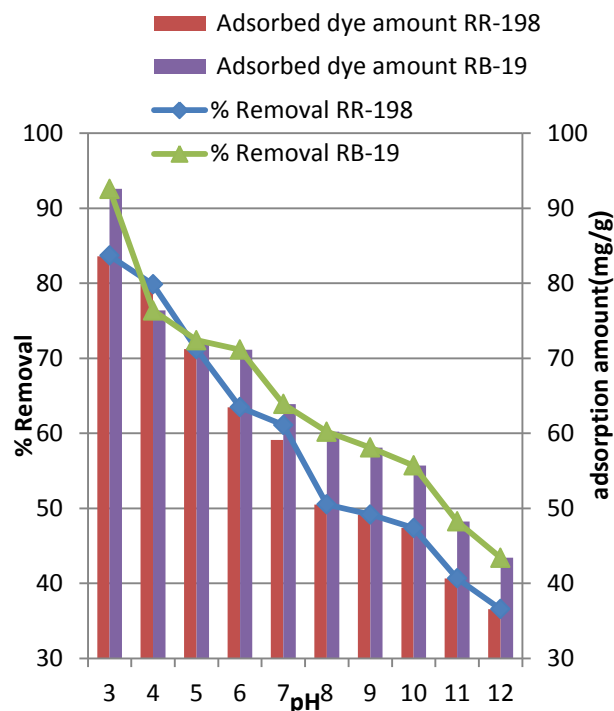


Fig.1. Effect of pH on color removal efficiency and adsorption concentration (Concentration of color 40 mg/L, contact time of 30 min, adsorbent dosage 0.4 g/L, temperature 35°C)

Sulak *et al.* [26] observed that the amount of dye adsorbed per unit weight of adsorbent decreased with increase in adsorbent mass. The amount of dye adsorbed decreased from 11.64 to 5.00 mg/g for an increase in adsorbent concentration from 2 to 8 g/L, whereas the percentage color removal increased from 48.26% to 79.92% with an increase in adsorbent concentration from 2 to 8 g/l. They the decrease in the amount of dye adsorbed with increasing adsorbent mass was due to adsorption sites remaining unsaturated during the adsorption reaction. This was due to the fact that as the dosage of adsorbent was increased, there was a less commensurate increase in adsorption resulting from the lower adsorptive capacity utilization of the adsorbent. Bulut *et al.* [27] reported that the sorption of Direct Blue 71 by wheat shells from aqueous solution occurred at low biosorbent doses. Similar behaviour regarding the absorption of anionic dyes on peanut hull was observed by Gong *et al.* [28].

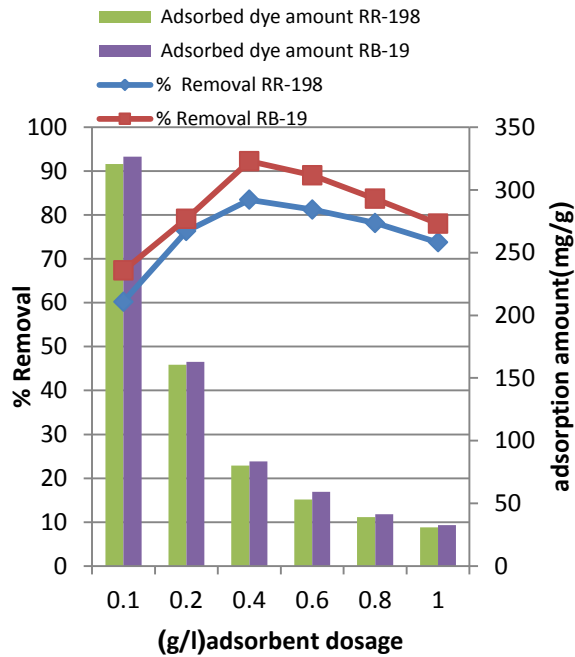


Fig.2: Effect of adsorbent dosage on dye removal efficiency and adsorption concentration (Concentration of color 40 mg/L, contact time of 30 min, pH =3, temperature 35°C)

Effect of Contact Time

The known of the equilibrium contact time are important for designing adsorption process [29]. Adsorption studies were carried out for 60 min in order to determine the effect of contact time on the percentage removal of RR-198 and RB-19 dyes as shown in Fig. 3. Dye adsorption on to Aloe Vera leave ash increased with time. For the 20 min contact time, 0.4 g of ash removed the RR-198 to 81.41% and RB-19 91.18% for initial dye concentration of 40 mg/l solution and at 35°C. The adsorption capacity of the ash of Aloe Vera leave was slightly greater for the RB-19 than that of the RR-198. After the equilibrium time (20 min) slight dye adsorbed by ash of Aloe Vera leave. Amin investigated the effect of contact time on the percentage removal of Reactive orange (RO) dye at different initial dye concentration [2]. The percentage removal of dye by different activated carbons was rapid in the beginning, but it gradually decreased with time until it reached equilibrium. The rate of removal is higher in the beginning due to larger surface area available of adsorbent. After adsorption, the rate of dye uptake is controlled by the rate of dye transported from the exterior to the interior sites of the adsorbent particles [30]. Also El Nemr *et al* observed that more than 64%

removal of DB-86 concentration occurred in the first 5 min [31].

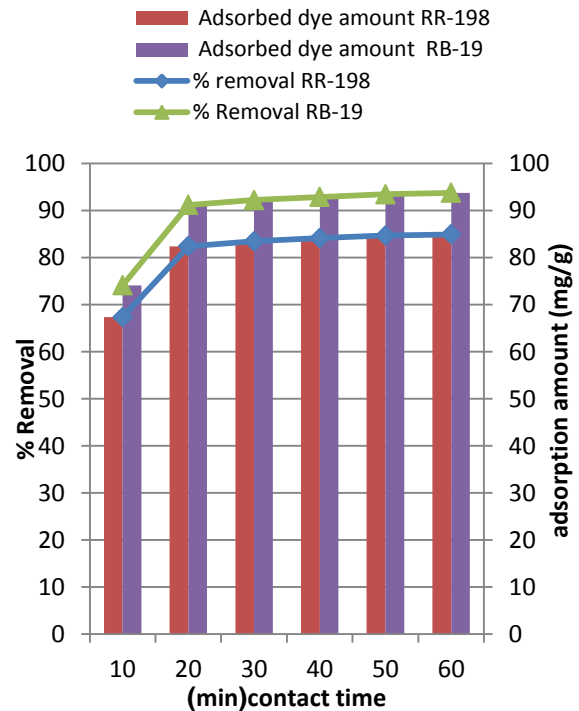


Fig.3: Effect of contact time on dye removal efficiency and adsorption concentration (Concentration of color 40 mg/L, adsorbent dosage 0.4 g/L, pH =3, temperature 35°C)

Effect of Initial Dye Concentration

The concentration of the dye in solution can strongly affect the sorption kinetics. Fig. 4 showed the effect of initial dye concentration (C_0) (10–160 mg/L) on the adsorption capacity and removal efficiency by the ash of Aloe Vera leave. The increase in initial dye concentration from 10 to 60 mg/L, causes an increase in the amount of dye adsorbed on the adsorbent. The effect of C_0 observed in this study suggests that the increase in C_0 enhances the interaction between dye and adsorbent [32]. In another word, increasing in the initial dye concentrations cause to increase in the driving force to overcome the resistance of the mass transfer of dye between the solution and the adsorbent surface [33]. At high initial Reactive RR-198 and RB-19 concentrations, the driving force favours adsorption because the gradient between the dye solution and the center of the particle enhances the dye diffusion through the film surrounding the particle. The concentration of the dye in the solution can also affect strongly the mechanism controlling the overall kinetic

coefficient. Also, it might be due to unsaturation of binding sites of the biomass, which resulted in an increase in the dye removal [34]. Bulut *et al.* [27] observed that the amount of dye sorbed per unit mass of biosorbent increases with increase in initial dye concentration from 50 to 250 mg/L. Khaled *et al.* [35] investigated the effect of initial concentration of Direct N Blue-106 on the biosorption of orange peel carbon. The amount of dye adsorb, q_e , increased from 20.42 to 54.39, 11.36 to 28.34 and 7.99 to 23.09 mg/g with an increase in the dye concentration from 50 to 150 mg/L at biosorbent doses 2, 4 and 6 g/L, respectively. Using Chitosan, Gibbs *et al.* [36] observed that, in the concentration range 50–100 mg/L, Reactive Black 5 concentration hardly influenced the time required to reach equilibrium. Similarly the biosorption of RR-198 and RB-19 on ash of Aloe Vera leave increased with an increase in dyes initial concentrations, suggesting that available sites on the biosorbent are the limiting factor for dye removal. Maximum removal (118.39 mg/g for RR-198 and 135.63 mg/g for RB-19) was observed with initial dye concentration 60 mg/L, biosorbent dose 0.4 g/L and pH 3, contact time 20 min, and temperature 35°C. Also was observed a limited effect of the dye concentration on sorption kinetics of RR-198 and RB-19 using Aloe Vera leave ash in the dye concentration up to 60 mg/L. Safa *et al.* [37] observed that maximum removal of Ever direct Orange-3GL (27.72 mg/g) with initial dye concentration 125 mg/L, biosorbent dose 0.05 g/L and pH 1 and Similarly for Direct Blue-67 dye were initial dye concentration 125 mg/L, biosorbent dose 0.1 g/L and pH 3.

Effect of Temperature

Fig. 5 show the effect of temperature on the adsorption kinetics of the Aloe Vera leave ash at pH 3 and 60 mg/L initial dye concentration. The temperature range used was from 25 to 55°C. Adsorption is favoured by increase in temperature from 25 to 55°C. The maximum adsorption capacity of the dye increased from 47.27 for RR-198 and 54.32 for RB-19 at 25°C to 81.15 and 88.63 mg/g for RR-198 and RB-19 respectively at 55°C. This in an unexpected behaviour, but it has been reported by some authors when studying the adsorption of different type of dyes and other organic compounds over several adsorbents ozerGok *et al.* [14], Ponnusami *et al.* [38], Ravikumar *et al.* [39], Hassan *et al.* [40]. ozerGok *et al.* [31] suggested that this behaviour is due to

the possibility of an increase in the porosity and total pore volume of the adsorbent with the increase of the temperature.

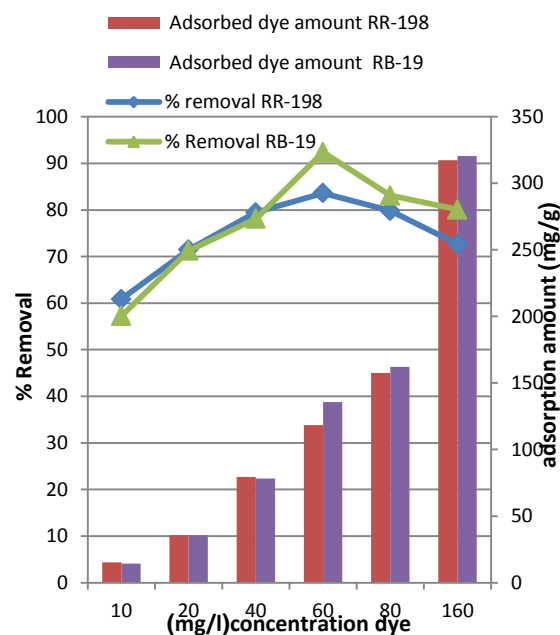


Fig.4: Removal percentage and adsorption concentration of dye differences concentrations (contact time of 20 min, adsorbent dosage 0.4 g/L, pH =3, temperature 35°C) Ravikumar *et al.* [39] suggested that increase in temperature produce swelling of the internal structure of the adsorbent causing increased penetration of the larger dye molecules. The increase of adsorption with temperature could be due to the enlargement of the pore sizes of adsorbent particles at elevated temperatures [8]. Results are shown that temperature plays an important role in the dye adsorption capacity of ash of Aloe Vera leave. This indicates that adsorption of RR-198 and RB-19 on to Aloe Vera leave ash is controlled by endothermic processes.

Adsorption Isotherms

Langmuir and Freundlich isotherm are frequently used to describe the equilibrium behavior of an adsorption process [2,41]. The Langmuir isotherm was given by the following equation:

$$\frac{C_e}{q_e} = \frac{1}{(Q_0 K_L)} + \left(\frac{1}{Q_0}\right) C \quad (2)$$

Where q_e (mg/g) and C_e (mg/L) are the amounts of adsorbed dye per unit weight of adsorbent and equilibrium dye concentration in solution, respectively. Q_0 (mg/g) is a maximum adsorption capacity of the dye (forming a monolayer) per unit weight of adsorbent. K_L is a constant related to the affinity of the binding sites (L/mg).

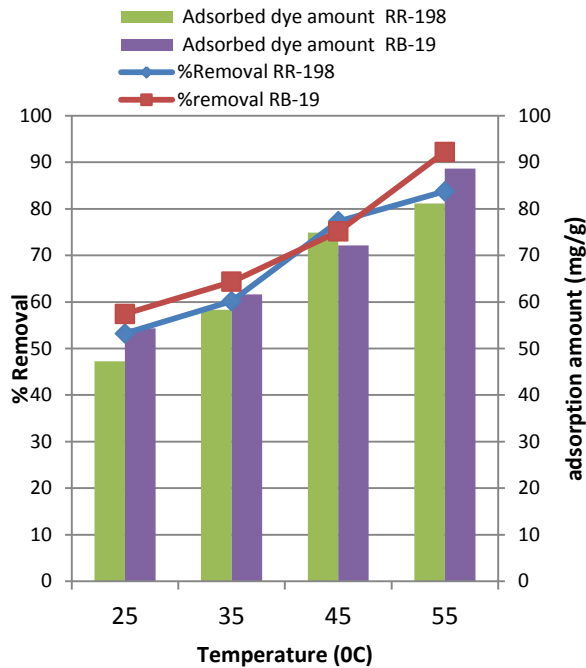


Fig.5: Effect of temperature on dye removal efficiency and adsorption concentration (Concentration of color 60mg/L, adsorbent dosage 0.4g/L, pH =3, contact time of 20 min)

The essential characteristics of the Langmuir isotherm can be expressed by a separation factor R_L , which is defined by the following equation [6]:

$$R_L = \frac{1}{(1 + K_L C_0)} \quad (3)$$

Where C_0 (mg/L) is the initial concentration of dye. R_L indicates the nature of the adsorption process to be unfavourable ($R_L > 1$), linear ($R_L = 1$), favourable ($0 < R_L < 1$), and irreversible ($R_L = 0$). The empirical Freundlich equation was given as follows [7]:

$$\ln q_e = \ln K_f + \frac{1}{n} \ln C_e \quad (4)$$

Where K_f and n are constants indicating adsorption capacity and intensity, respectively. The Freundlich adsorption constant, n , should be in a range of 1–10 for beneficial adsorption. Table 3 showed the Q_0 and K_L values for the Langmuir isotherm, the K_f and n values for the Freundlich isotherm, and the regression coefficients (r^2) obtained from the linear regression equation between the values of C_e/q_e and C_e and $\ln q_e$ and $\ln C_e$, respectively (Fig. 6 and 7). For the both days, the high regression coefficients for the Freundlich and Langmuir isotherms (for RR-198: $r^2=0.99$ and RB-19: $r^2=0.91$) indicated that the Freundlich model were good fit with the experimental data. The Q_0 from the

Langmuir isotherm indicated that the adsorption capacity of ash of Aloe Vera leave was greater for the RR-198 (334.67 mg/g) than the RB-19 (29.81 mg/g). The R_L values of the RR-198 and RB-19 were found to be between 0.008 to 0.58 and 0.047 to 0.76 for dye concentrations of 10–160 mg/L, respectively. The observed R_L values indicate favorable adsorption of the RR-198 and RB-19 on fly ash ($0 < R_L < 1$).

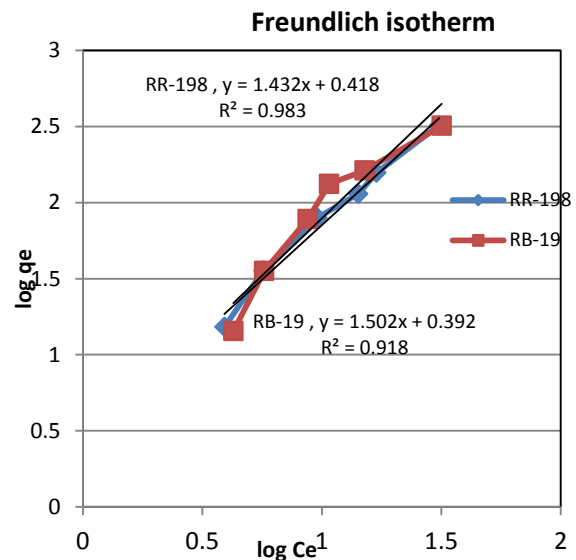


Fig.6.: Evaluating the Freundlich isotherm in eliminating dyes by ash of Aloe Vera leave

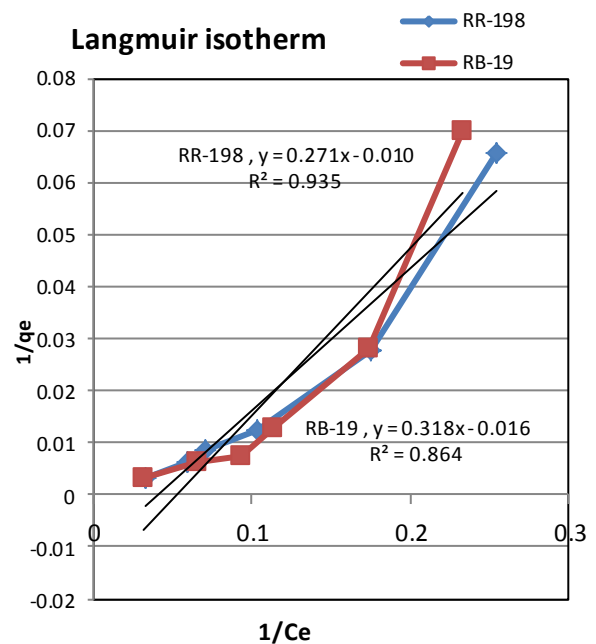


Fig.7. Evaluating the Langmuir isotherm in eliminating dyes by ash of Aloe Vera leave

Table 2. : The Q_0 and K_L values for the Langmuir isotherm, the K_f and n values for the Freundlich isotherm and the regression coefficients of equations

Dye	Langmuir			Freundlich		
	Q_0 (mg/g)	K_L (1/mg)	r^2	K_f	n	r^2
Reactive Red198	34.67	0.285	0.935	2.01	6.21	0.983
Reactive Blue19	29.81	0.019	0.864	0.082	3.37	0.918

CONCLUSION

The experimental conditions of maximum adsorption of the dye were optimized. It was observed that under the optimized conditions, up to 80% for RR-198 and 90% for RB-19 of dyes can be removed from the solution onto the ash of Aloe Vera leave. Ash shows significant adsorption capacity for RR-reactive198 and RB-19 dyes under suitable experimental conditions and hence will serve as a useful adsorbent. The ash of Aloe Vera leave is abundantly available and inexpensive too. Its binding capacity of reactive dye is appreciably higher. The method may be helpful for designing and fabricating a reactive dye rich effluent treatment plant in the future.

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