# Utilization of Peanut (*Arachis hypogaea*) Hull Based Activated Carbon for the Removal of Amaranth Dye from Aqueous Solutions

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**ABSTRACT:** *This research work is concerned with the investigation of removing amaranth foodstuff dye on activated carbon derived from peanut hulls (ACPH) as a low-cost adsorbent. The influence of different reaction parameters affecting dye uptake including pH, shaking speed, particle size, temperature, initial dye concentration, and contact time was investigated for proper selection of the optimized parameters for the removal process. By using ACPH, 76.92 mg/g adsorption capacity was achieved under agitation time of 90 min at pH 4 and temperature 60°C. Results demonstrate that experimental equilibrium data were fitted to Langmuir isotherm to a greater extent than Freundlich isotherm. Kinetics of amaranth dye on peanut hulls was found to follow pseudo-second-order kinetics. Results indicated that ACPH is a good adsorbent for removing amaranth dye from wastewater.*

**KEYWORDS:** *Amaranth; Peanut hull; Activated carbon; Kinetic modeling.*

## INTRODUCTION

Most industries, especially leather, paper, textile, food, cosmetic and plastic use dyes to color their products [1, 2]. The dyes (soluble or insoluble) are generally synthetic in nature, having intricate molecular structures and embodied with different functional groups that make them stable and tough to biodegrade [3]. Dyeing process in textile industries is one of the basic causes of contamination being participating in environmental pollution. Annually, around 10,000 different types of dyes mostly azo dyes and pigments are produced worldwide [4]. Dyes concentration in textile industries ranges from 10-10,000 ppm depended upon the dye removing treatments [5]. Wastewater discharged by several industries e.g. textile industry, contains amaranth dye that is undesirable for environment due to its offensive side effects [2, 6-8]. A variety of products requires the use of amaranth dye in manufacturing process and can be found in the effluent of the majority sewage treatment services. There are a number of methods which have been developed to remove various health hazardous chemicals from waste material like organic and inorganic impurities [9]. These include: reverse osmosis [10, 11], biological treatment, advanced oxidation processes, coagulation [12, 13] and flocculation, photocatalytic process etc. [14]. Treatment by using these methods showed color reduction in dye solution but these are expensive method for decolonization.

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A search for more effective method is important because of economical reasons and adsorption is considered to be an effective and economical strategy for dyes and other pollutants removal from wastewater [13, 15-23]. The chief adsorbent that is used in industry for adsorption purpose is the activated carbon prepared by using various sources [24]. Powdered peanut (*Arachis hypogaea)* hulls are used as activated carbon for the removal of dyes because nuts of agricultural wastes are available locally, economical [25, 26] and accessible from the 3500 acreage following yearly production of 140,000 tons of peanuts in Pakistan. Peanut hulls are generally available in markets as waste material so these are used to remove amaranth dye from wastewater. The present investigation reports a simple method for removal of amaranth dye by activated carbon derived from peanut hulls. For this purpose, various factors affecting the biosorption, such as pH of solution, shaking speed, particle size, temperature and contact time were investigated by the batch equilibration technique.

## EXPERIMENTAL SECTION

### *Chemicals and instruments*

Sodium hydroxide (NaOH), hydrochloric acid (HCl) and phosphoric acid ($H_3PO_4$) were purchased from Sigma (Sigma-Aldrich, Taufkirchen, Germany). Amaranth dye having molecular weight of 604.473 g/mol was purchased from Merck (Merck Damstadt, Germany) and its structure is given in Fig. 1.

A pH meter (inoLab pH 720, WTW, Weilheim, Germany) was employed for the pH measurements. The spectrophotometric measurements were carried out with a UV-Vis spectrophotometer (UV 4000, MRI, Germany). The solutions were stirred on orbital shaker **(**Wisd, WiseShake SHO-2D, Seoul, South Korea**).**

### *Preparation of adsorbent*

Peanut (*Arachis hypogaea)* hulls were collected from local market of Kotla Arab Ali Khan (Pakistan) and dried in air. Fine powder of peanut hulls was prepared by grinding. In order to convert it into activated carbon, 10 g of this fine powder was taken and 20 ml of $H_3PO_4$ was added into it followed by stirring at 105°C. After that the mixture was heated in furnace at 405°C for 2 hours and after washing it was allowed to dry in oven overnight [27]. This activated carbon derived from peanut hulls (ACPH) was used as adsorbent for removal of amaranth dye.

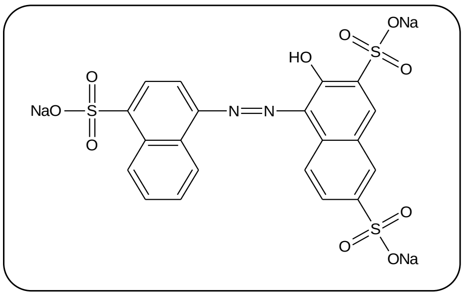


***Fig. 1: Chemical structure of amaranth dye.***

### *Equilibrium adsorption studies*

Adsorption study was performed to find the effect of various factors on the adsorption behavior of amaranth dye onto ACPH. Different factors that control adsorption such as pH, stirring speed, particle size, temperature and contact time were optimized while studying the adsorption isotherm and kinetics of the reactions. Dye solutions with known concentrations were prepared and required amount of ACPH was added followed by shaking on orbital shaker for predefined time. Then the solutions were filtered and residual concentration of amaranth dye in the filtrate was determined by monitoring the absorbance at 521 nm using UV-Vis spectrophotometer. Adsorption capacity and percentage removal were calculated according to following equations 1 and 2 respectively [28].

$$q_e = (C_o - C_e) \times \frac{V}{W} \tag{1}$$

$$\%\ \text{of dye removal} = \frac{C_o - C_e}{C_o} \times 100 \tag{2}$$

Where, $C_o$ is the initial concentration (mg/L), $C_e$ is the equilibrium concentration (mg/L) of dye, $V$ is the volume (L) and $w$ is the weight of adsorbent (g).

## RESULTS AND DISCUSSION

### *Effect of different parameters on adsorption of amaranth dye on ACPH*

The pH of the solution is found to have significant effect on the properties of the adsorbate and adsorbent. Influence of pH on the adsorption of amaranth dye was studied by varying the pH from 3-11 by keeping all the other factors constant. The results are shown in Fig. 2A.

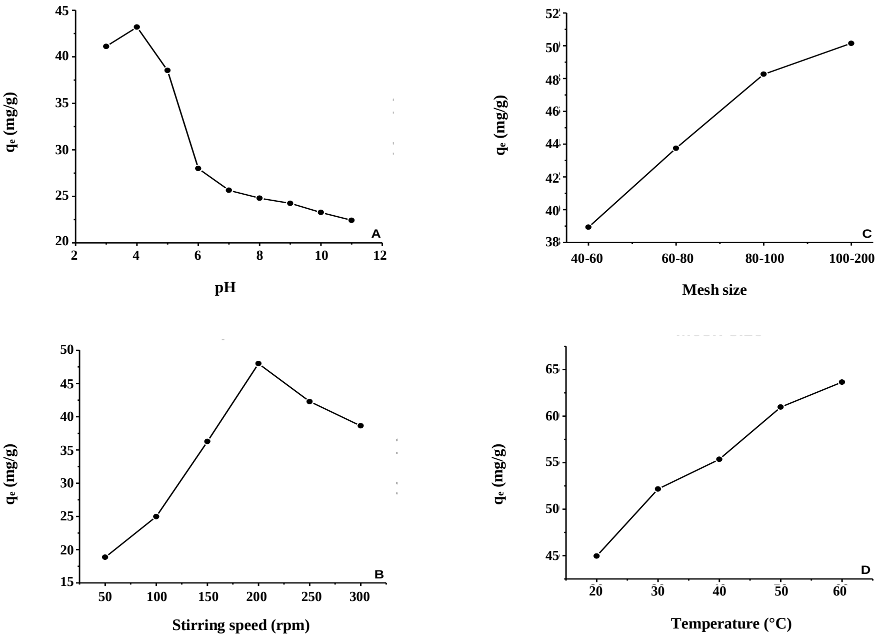


*Fig. 2: Effect of the solution pH (A), stirring speed (B), adsorbent particle size (C), temperature (D) on the adsorption capacity of amaranth dye onto ACPH.*

From the graph one can see that the maximum adsorption of dye was found at pH 4 and further increase in pH results in decrease in adsorption capacity. The point of zero charge ($pH_{PZC}$) of the adsorbent was found to be 6. The adsorbent potential surface became neutral, as pH of aqueous solution was equal to $pH_{PZC}$ and is negatively charged when $pH > pH_{PZC}$ while it gets positively charged when $pH < pH_{PZC}$. The electrostatic attraction between the dye molecules (negatively charged) and ACPH surface (positively charged; $pH_{PZC} = 6$) might be the predominant adsorption mechanism at acidic pH. The increase in pH value caused a decrease in positive charge of the ACPH surface. This decreased the electrostatic forces among ACPH and amaranth dye, so there is reduction in adsorption of dye as the pH increased from 5 to 11.

The influence of stirring speed on adsorption process was studied by varying the stirring speed from 50 to 300 rpm at pH 4 and results are shown in Fig. 2B. The results showed that the adsorption capacity of amaranth dye was increased upto 200 rpm, when shaking speed increased from 50 to 300 rpm. The possible explanation of this increase could be that as the shaking speed increased, the collision between dye and adsorbent surface was also increased which results in fast reaction between positively charged ACPH and anionic dye. At higher shaking speed a decrease in adsorption capacity of dye was observed.

Particle size of adsorbent affects the adsorption process, as smaller particle size would have greater proficiency to adsorb dye due to its larger surface area. The ratio of dye adsorbed approached the maximum value when the sorbent particle size was very small. Different mesh sizes from 40 to 200 microns were prepared from ACPH and influence of mesh size was studies at pH 4 and stirring speed 200 rpm. The results presented in Fig. 2C showed that the highest adsorption capacity was achieved by 100–200 mesh size. For convenience the mesh size 80–100 was used in further experiments.

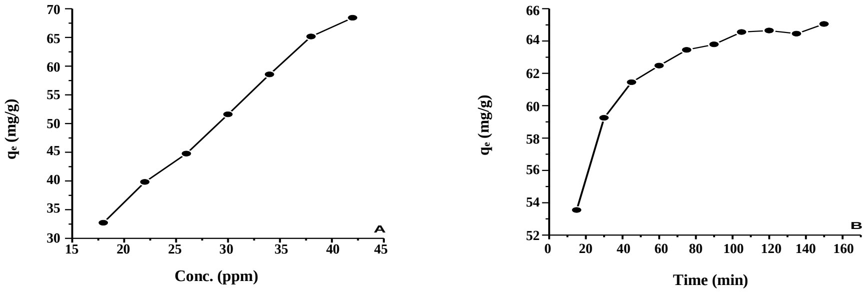


***Fig. 3: Effect of initial dye concentration (A) and contact time (B) on the adsorption capacity of amaranth dye onto ACPH.***

The effect of temperature was studied by varying the temperature from 20°C to 60°C at pH 4, stirring speed 200 rpm and mesh size 80–100. The obtained results (Fig. 2D) showed that the increase in the temperature of the solutions of amaranth dye from 20 to 60°C leads to an increase in the adsorption capacity of ACPH, which shows that the adsorption process is endothermic and chemical in nature. The possible explanation of this increase in adsorption capacity of amaranth dye onto ACPH could be due to the availability of more active sites and activation of the adsorbent surface at higher temperatures and increased diffusion and mobility of amaranth dye ions from the bulk solution towards the ACPH surface.

The apparent capacity of ACPH for amaranth dye was determined at the different concentrations. Fig. 3A clarifies the relation between capacities and the dye ion concentrations, which shows that as the dye ion concentration increased the adsorption capacity increased until 61.50 mg/g at 42 ppm. In general, the data indicated that sorption capacity increased with increase in initial dye ion concentration on ACPH. This sorption characteristic indicated that surface saturation was dependent on the initial dye ion concentrations. At low concentrations, adsorption sites took up the available dye more quickly. However, at higher concentrations, dye needed to diffuse to the biomass surface by intraparticle diffusion and greatly hydrolyzed ions will diffuse at a slower rate. The maximum dye ion sorption of an adsorbent may be determined from column experiments, by the use of a large excess of the adsorbate.

The contact time has a significant effect on adsorption process. The adsorption capacity of amaranth dye was determined by varying the contact time from 15–150 min at pH 4, stirring speed 200 rpm, mesh size 80–100 and temperature 60°C. The results are shown in Fig. 3B. The rate of dye adsorption was rapid in the first 30 min and equilibrium was established in 80 min where adsorption efficiency reached its maximum value. After that no further increase in adsorption capacity was observed. At start more adsorption occurred due to availability of vacant sites on the surface of adsorbent but after that saturation occurred and adsorbate molecules could not enter in the inner sites due to high degree of saturation.

***Equilibrium modeling***

The equilibrium data of amaranth dye were analyzed by fitting into Langmuir and Freundlich equations to find out the suitable model that may be used for design consideration. Table 1 summarizes the constants and coefficients of different models. The linear form of Langmuir isotherm is given by the following equation [29]:

$$\frac{1}{q_e} = \frac{1}{q_m} + \frac{1}{K_L q_m} \times \frac{1}{C_e} \tag{3}$$

Where $q_m$ is the maximum adsorption capacity relative to complete monolayer capacity (mg/g), $K_L$ is Langmuir constant. Values of $q_m$ and $K_L$ are determined from the linear regression plot of $1/C_e$ and $1/q_e$ (Fig. 4A). Linear plot depicts the Langmuir adsorption plot reveling the preferential monolayer formation.

The characteristics of Langmuir isotherm can be expressed by a dimensionless constant, known as separation factor ($R_L$), which can be represented as:

$$R_L = \frac{1}{1 + K_L C_o} \tag{4}$$

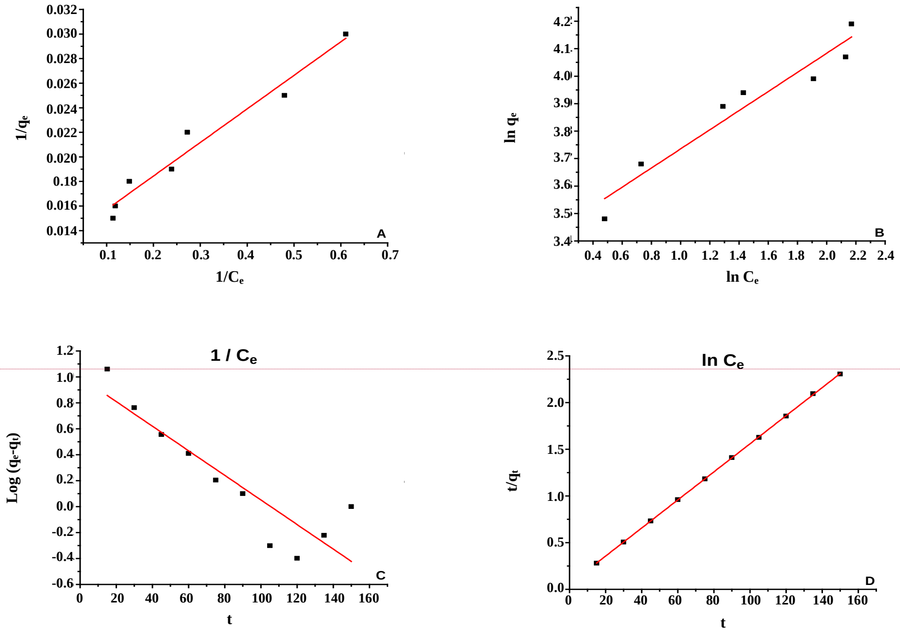


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*Fig. 4: Langmuir isotherm model (A), Freundlich isotherm model (B), Pseudo first order kinetic model (C) and Pseudo second order kinetic model (D) for amaranth dye removal using ACPH.*

$R_L$ value is an indication of adsorption nature to be unfavorable ($R_L > 1$), linear ($R_L = 1$), favorable ($0 < R_L < 1$) or irreversible ($R_L = 0$). $R_L$ value for the adsorption of amaranth dye is found to be 0.047, which shows that adsorption is favorable.

The Freundlich isotherm involves multilayer formation on the surface of heterogeneous systems. The linear form of Freundlich equation is given as [30]:

$$\ln q_e = \ln K_F + \frac{1}{n} \ln C_E \quad (5)$$

Where $K_f$: is the distribution coefficient tells the amount of dye adsorbed on adsorbent for unit equilibrium concentration, n is the adsorption intensity. Values of $K_F$ and $n$ are determined from the linear regression plot of ln $q_e$ versus ln $C_e$ (Fig. 4B). The value of n is found to be more than 1 and the value of 1/n is lie between ($0 < 1/n < 1$) so attributed to cooperative adsorption of dye and revealed chemisorption process. The $R^2$ value of Freundlich model was lower than that obtained from Langmuir isotherm and also the calculated maximum adsorption capacity ($q_m$) from Langmuir isotherm agree well with experimental adsorption capacity showing that the equilibrium data best fitted to Langmuir Isotherm than Freundlich isotherm model which suggest monolayer adsorption of amaranth dye on the ACPH.

### *Kinetic modeling*

Kinetics analysis for adsorption process can predict the rate at which a pollutant is removed from aqueous solutions and provides valuable data for understanding the mechanism of adsorption process. Pseudo first order and pseudo second order kinetic models are used to evaluate the mechanism involved in the process of adsorption. A linear form of pseudo first order equation is given as [31]:

***Table 1: Adsorption isotherms parameters and adsorption kinetics constants of amaranth on ACPH.***

| Isotherms | Parameters | | Values |
|---|---|---|---|
| Langmuir | $R_L$ | | 0.047 |
| | $K_L$ (L/g) | | 0.481 |
| | $q_m$ (mg/g) | | 76.92 |
| | $R^2$ | | 0.956 |
| Experimental adsorption capacity | $q_e$ (mg/g) | | 68.42 |
| Freundlich | $K_F$ (mg/g) | | 29.55 |
| | n | | 2.870 |
| | $R^2$ | | 0.922 |
| Pseudo first order | $k_1$ (1/min) | | -0.022 |
| | $q_e$ (mg/g) | | 9.980 |
| | $R^2$ | | 0.784 |
| Experimental adsorption capacity | $q_e$ (mg/g) | | 65.05 |
| Pseudo second order | $k_2$ (g/mg.min) | | $4.06\times10^{-3}$ |
| | $q_e$ (mg/g) | | 66.53 |
| | $R^2$ | | 0.999 |
| Thermodynamic | ΔH (kJ/mol) | | 18.50 |
| | ΔS (J/mol.K) | | 42.55 |
| | ΔG° (kJ/mol) | 20°C | -2.450 |
| | | 30°C | -4.870 |
| | | 40°C | -6.120 |
| | | 50°C | -8.340 |
| | | 60°C | -11.45 |

$$\log(q_e - q_t) = \log q_e - \frac{k_1}{2.303}t \qquad (6)$$

Where $q_t$ is the amounts of adsorbed dye (mg/g) at time t and $k_1$ is the pseudo first order rate constant. Values of $q_e$ and $k_1$ are calculated from the plot of log ($q_e - q_t$) versus t (Fig. 4C). The value of the correlation factor $R^2 = 0.784$, and the value of theoretical $q_e$ is greatly differ from the experimental value suggesting that pseudo-first order model not explain the kinetics of adsorption (Table 1).

The linear form of pseudo second order model is given in Eq. (7) [32, 33]:

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \qquad (7)$$

Where $k_2$ is the pseudo second order rate constant. A plot between t and $t/q_e$ was used to determine the values of $q_e$ and $k_2$ (Fig. 4D). The value of correlation coefficient $R^2 = 0.999$ which is higher than that obtained from pseudo first order model. In addition, the theoretical $q_e$ value agrees with the experimental $q_e$ value. The results obtained reflect that the adsorption of amaranth dye precedes second order kinetics mechanism. The values of constants and coefficients of different models are given in Table 1.

***Thermodynamic modeling***

Thermodynamic parameters such as, free energy change (ΔG°), enthalpy (ΔH) and entropy (ΔS) were evaluated to confirm the nature of adsorption of amaranth dye onto ACPH and can be estimated from the following equations:

$$\Delta G^o = -RT \ln K_d \qquad (8)$$

$$\ln K_d = -\frac{\Delta H}{RT} + \frac{\Delta S}{R} \qquad (9)$$

Where $K_d$ is the thermodynamic equilibrium constant. The values of $\Delta H$ and $\Delta S$ were calculated from the slope and intercept by plotting ln $K_d$ versus 1/T using

***Table 2: Comparison of amaranth dye adsorption using ACPH with other reported adsorbents.***

| Adsorbent | Adsorption capacity (mg/g) | Reference |
|---|---|---|
| *Arachis hypogaea* shells | 10.53 | [34] |
| Powered peanut hull | 14.90 | [24] |
| Potato peels | 1.710 | [35] |
| Bottom ash | 7.860 | [36] |
| De-Oiled soya | 12.70 | [36] |
| $MgAlCO_3$ | 120.9 | [37] |
| Water hyacinth leaves | 70.00 | [38] |
| Core/Shell nanocomposites | 142.1 | [39] |
| ACPH | 76.92 | This study |

equation 9 and the values of ΔG° were calculated according to equation 8.

Table 1 summarizes the values of ΔG°, ΔH and ΔS. The positive values of ΔH and ΔS show that the adsorption process is endothermic with increasing the randomness of the system and dominant by chemisorption. The negative value of free energy indicates that the adsorption process is spontaneous.

Table 2 showed the comparison of amaranth dye's theoretical adsorption capacity onto ACPH with other reported adsorbents. It can be perceived that the ACPH has a comparable and good adsorption capacity, which showed that ACPH could be suitable adsorbent for azo dyes removal.

## CONCLUSIONS

Activated carbon derived from waste material powdered peanut hulls were effectively utilized, as adsorbent for the removal of amaranth dye from aqueous solution, since the peanut hulls were easily available in large quantity in Pakistan and the treatment method seemed to be economical. The maximum adsorption capacity of amaranth dye onto ACPH was achieved at initial pH 4, stirring speed 200 rpm, mesh size 80-100 and temperature 60°C. Isothermal data of adsorption process followed Langmuir model. The kinetics study clearly demonstrates that adsorption follow pseudo-second-order kinetics. The adsorption capacity was found to be 76.92 mg dye per gram of the adsorbent for amaranth. Thus adsorption can be a feasible for wastewater treatment and ACPH can act as a good and economical adsorbent.

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