

Original Research Article

Kinetics and thermodynamics of enhanced adsorption of E120 dye using activated carbon

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Abstract

Purpose: To study the adsorption of dye (E120) from aqueous solution onto activated carbon.

Method: Factors influencing adsorption were examined and optimized. Three adsorption isotherm models (Langmuir, Freundlich and Temkin) were investigated. Agitation time was set at 72 hours, E120 dye concentration at 10 – 80 mg/L, pH at 7, temperature at 25 °C and mass at 125 mg.

Results: Adsorption of E120 dye onto activated carbon was enhanced by decreasing the mass of activated carbon, pH and ionic strength of the solution and by increasing the temperature. Under optimal conditions, the maximum adsorption capacity of activated carbon for E120 dye was 10.1 mg/g at 30 °C. The model parameters were 0.307 L/mg (K_L), 10.1 mg/g (q_m), 0.9491 (R^2) for the Langmuir isotherm; 2.98 (n), 0.445 mg/g (K_f), and 0.6592 (R^2) for Freundlich isotherm; and 4.59 mg/L (A), 2.23 J/mol (B), and 0.5914 (R^2) for Temkin isotherm. Thermodynamic studies indicate that the adsorption of E120 dye onto activated carbon is an endothermic process with an adsorption enthalpy (ΔH) of 8.7 KJ/mol. The positive values for ΔG indicate that adsorption was non-spontaneous. The kinetic study of E120 dye adsorption showed that the adsorption process obeyed pseudo-second order kinetics.

Conclusion: Commercially available activated carbon, in terms of its physical and chemical characteristics, is a superior adsorbent to other adsorbents mentioned in the literature for removal of toxic dye E120 from aqueous solutions at a high removal capacity.

Keywords: Carmine dye E120, Activated carbon, Thermodynamics, Adsorption isotherm, Kinetic models

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INTRODUCTION

Adsorption is a simple, economic and highly efficient physicochemical process that can be used to optimize the treatment of waste, understand clarification and depollution of industrial liquids [1]. Applications of various adsorption methodologies have expanded rapidly

due to rising environmental and quality requirements. They have been used for treating aqueous solutions to remove highly toxic dissolved organic dye components [2]. During the synthesis and application of dyes, large quantities of untreated dyes are discharged into the environment. An example of such dye is carmine (a red azo dye) also known as natural red E120 (Figure 1) [3].

E120 is present as an additive in many products such as artificial flowers, rouge, cosmetics, and some medications [4]. It is also one of the most common dyes used in textile, juice, medical and pharmaceutical applications [5]. Except in low concentrations when used in applications involving human contact or ingestion, E120 is highly toxic causing skin, and eye related diseases, cancer and is neuro-toxic [6]. According to the World Health Organization (WHO), E120 is considered safe at concentrations lower than 0.005 mg/L [7]. Several methods have been used to remove carmine dye from aqueous solutions including chemical precipitation, ultra-filtration, electrochemical deposition and coagulation-flocculation. However, these methods are expensive and have variable efficiency in removing carmine dye from aqueous solutions [8].

Activated carbon is a microporous material used in many applications as adsorbent for removal of dye pollutants from aqueous solution [9]. In this work, the adsorption of carmine dye from aqueous solution using activated carbon was studied as a model system to demonstrate the ability of activated carbon to remove this carcinogenic material from aqueous solution. The effects of different experimental conditions were also investigated.

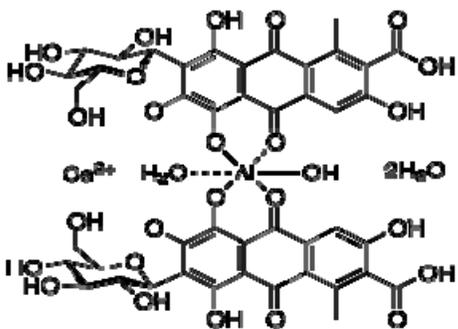


Figure 1: Chemical structure of carmine dye (E 120)

EXPERIMENTAL

Materials

All reagents used in this work were of analytical grade and were used as received without any further purification. The E120 dye was obtained from Lobal Chemie (India) and activated carbon with particle diameter ranging from 300 to 500 μm , surface area of 500 - 3000 m^2/g and a total pore volume of 1.4 cm^3/g was purchased from Nen Tech Ltd (UK). The physiochemical properties of activated carbon were reported by

Al-Degs et al [10]. Activated carbons have a large specific surface area (820 m^2/g) and various bulky functional groups, which give the activated carbon the ability to adsorb considerable amounts of E120 dye. The acidity constant (pK_a) of E120 dye, reported in the literature and not provided by the manufacturer is 5.43 [11].

Adsorption of E120 dye

The adsorption properties of the dye were studied using the batch method. A known mass of activated carbon (300 - 500 μm diameter) was added to 100 mL of the dye solution and the mixture was agitated for a known time. The final dye concentration in the solution was measured using a double beam UV-visible spectrophotometer (SP-3000 nano, Optima, Japan). The amount adsorbed by the activated carbon was calculated as the difference between the initial and final dye concentrations. The effects of carbon mass, dye concentration, solution pH, ionic strength, and temperature on E120 dye adsorption were investigated as outlined below.

The concentrations of E120 dye remaining in solution were determined spectrophotometrically, at 290 nm. A linear calibration curve was obtained using a series of standard samples in the concentration range of 10 - 80.0 mg/L. A linear calibration graph was obtained with correlation coefficient (R^2) = 0.9641. All experiments were conducted in triplicate. The uncertainty of the measurements was calculated from the standard deviation and estimated to be in all cases less than 1%.

Effect of agitation time on E120 dye

A mass (125 mg) of activated carbon was added to 100 mL solutions initially containing 80.0 mg/L of E120 dye and agitated for times varying from 24 to 120 hours at 25°C and pH 7. The concentration of the remaining dye was determined.

Effect of solution pH, ionic strength and mass of adsorbent on E120 dye adsorption

Carmine dye solution (100 mL) of initial concentration 80.0 mg/L was placed in a 100 mL volumetric flask. The flask was closed and agitated for 72 hours at 25°C, after which, the concentration of dye remaining was measured. Effect of solution pH (1.0, 2.0, 3.0, 7.0, 9.0, 10.0 and 11.0) was recorded using an Ezdo pH meter-pp-201 (Taiwan). The mass of activated carbon (25, 50, 75, 100, 125, 150, 175 and 200 mg) on

E120 dye adsorption were also recorded. The pH was monitored in the beginning and at the end of the experiments and was adjusted with either acid or base to the required pH. The effect of ionic strength on the adsorption of the carmine dye was determined by placing 125 mg of activated carbon in 100 mL of solutions containing 80.0 mg/L of carmine dye, with different concentrations of sodium chloride (0.1 to 0.5 mol/L) and the solutions were agitated for 72 h at 25°C and pH 7. The dye concentration remaining after agitation was determined spectrophotometrically.

Effect of E120 dye concentration

To investigate the effect of dye concentration, 125 mg of activated carbon was added to 100 mL of each of eight solutions, all at pH 7 but containing different concentrations of carmine dye (10.0 to 80.0 mg/L). The volumetric flasks were agitated for 72 hours at 25°C and the remaining dye concentration was determined.

Assessment of adsorption isotherms at different temperatures

A 125 mg activated carbon was added to each of eight solutions at pH 7 but containing different concentrations of E120 dye (10.0 to 80.0 mg/L). The volumetric flasks were agitated in a water-bath shaker at 25°C (298 K) for 72 h. The concentration of dissolved dye remaining was then determined, experiment was repeated at different water-bath temperatures (293 and 313 K). The concentration of adsorbed dye (q_e , mg/g) was calculated by the difference between the initial (C_0 , mg/L) and the equilibrium (C_e , mg/L) dye concentration. The adsorption of carmine dye was expressed in terms of the distribution coefficient K_d (L/g) as shown in Eq 2.

$$q_e = (C_0 - C_e) * V/m \dots\dots (1)$$

where q_e is the concentration of adsorbed dye in mg/g, C_0 is the initial concentration (mg/L), The equilibrium concentration C_e mg/L, V is the volume of the solution in L, and m is the mass of adsorbent (g).

$$(K_d) = q_e/C_e \dots\dots\dots (2)$$

RESULTS

The E120 dye contains a large number of functional groups. The physicochemical properties of E120 dye play a significant role in the effect of pH and ionic strength on the adsorption of E120 dye by activated carbon.

Effect of agitation time on E120 dye

Figure 2 shows that the K_d value for adsorption of carmine dye increased with time. A short contact time is desirable for economic reasons. K_d greater than 1 L/g is generally considered acceptable [12]. A contact time of 72 h, giving a K_d of 1.3 L/g, was chosen for subsequent experiments. It was found that 72 hours were sufficient to obtain a reasonable value of distribution coefficient (K_d) and this agitation time was used in subsequent experiments.

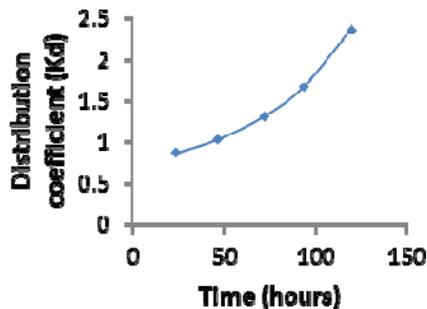


Figure 2: Effect of agitation time on E120 dye adsorption onto activated carbon

Effect of solution pH, ionic strength and mass of adsorbent on carmine dye

As shown in Figure 3, K_d increased with acidity of the solution, from 0.27 L/g at pH 11 to 2.96 L/g at pH 1. The adsorption of E120 dye on the activated carbon was strongly influenced by the pH of the solution. At low pH, the oxygen functional groups on the dye is protonated, as the pK_a was 5.3 and hydrogen bonds are formed between dye molecules and functional groups on activated carbon [13]. As the pH increases, the dye molecules become negatively charged and gets repelled by the negatively-charged activated carbon surface [14]. Furthermore, the increasing number of OH^- ions are strongly adsorbed and competes with dye molecules for adsorption sites [14]. Hydrophobic interactions between the aromatic part of the dye molecule (Figure 1) and the hydrophobic parts of the activated carbon surface are responsible for such binding of the dye molecules to the activated carbon [13].

As shown in Figure 5, distribution coefficient K_d value decreased from 3.71 L/g at 0.1 M NaCl to 0.23 L/g at 0.5 M NaCl. Thus, another one of the factors that play a role in adsorption of E120 dye from solution is the salt level in the solution. Adsorption of E120 dye decreased with increased NaCl concentration. The salt ions screen the surface of the activated carbon from the dye molecules and so reduce any attractive

electrostatic interaction between the dye and the activated carbon [13]. The added ions also compete with dye molecules for adsorption sites on the active carbon surface. Similar trend was reported by Boumediene *et al* [15]. They found that as the ionic strength increased, the adsorption capacity of the studied dye decreased. This was attributed to the competition between the ions and the dye on the adsorbent surface.

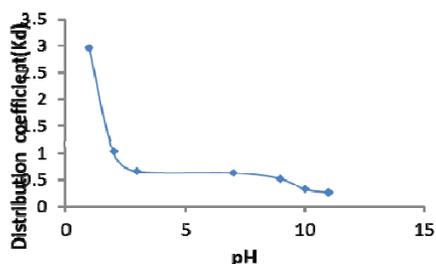


Figure 3: Effect of pH on E120 dye adsorption onto activated carbon

Removal of E120 dye from solution also depends on the amount of activated carbon per 100 ml of solution. As shown in Figure 4, K_d decreased when the mass of activated carbon increased. However, K_d values showed only small changes for masses greater than 125 mg (0.05 L/g).

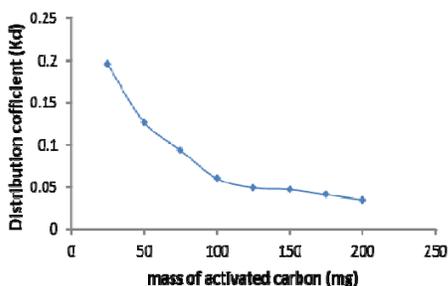


Figure 4: Effect of mass of activated carbon on E120 dye adsorption

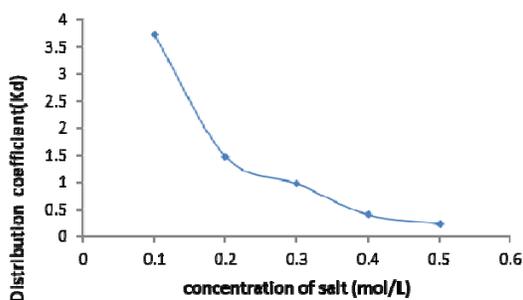


Figure 5: Effect of ionic strength on E120 dye adsorption

Effect of E120 dye concentration

The effect of concentration of E120 dye on its adsorption was investigated in the concentration range 10.0 - 80.0 mg/L, at pH 7, 25 °C, 72 h of shaking and activated carbon amount of 125 mg. As shown in Figure 6, the distribution coefficient (K_d) value increased when the concentration increased (K_d value increased from 0.49 L/g at 10 mg/L to 1.36 L/g at 80 mg/L) which might be due to increase in the available sites for adsorption of E120 dye. K_d changed to only a small extent with concentration above 60 mg/L which might be due to limited available sites of the activated carbon dose for the removal of E120 dye at higher concentration. Therefore 80 mg/L was selected as the initial concentration for investigating the effects of other variables.

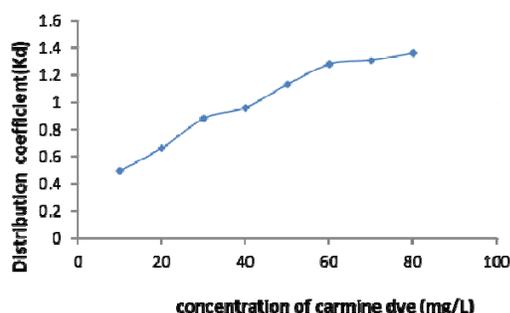


Figure 6: Effect of E120 dye concentration adsorption onto activated carbon

Adsorption isotherms

The effects of temperature on adsorption of E120 dye was measured using varying concentrations at different temperatures and the results are shown in Figure 7. The distribution coefficient of E120 dye increased with increasing temperature suggesting that this was an endothermic process. Higher temperatures may have led to the creation of new active sites on the activated carbon and thus permitted more adsorption of the dye, as suggested by Al-Degs *et al* [13].

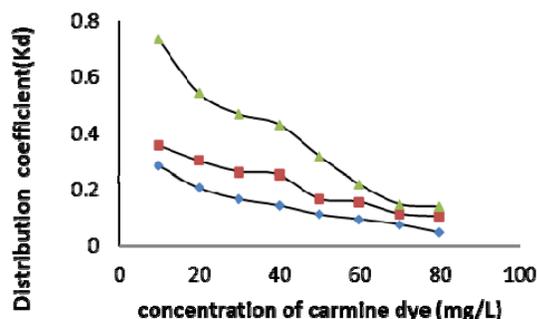


Figure 7: Effect of temperature on E120 dye adsorption onto activated carbon at 293 K (◆), 298 K (■), and 303 K (▲)

Langmuir, Temkin and Freundlich models were used to determine the maximum adsorption capacity of activated carbon for E120 dye. The fits of the adsorption data to the three models are presented in Figure 8. As shown in Table 1, the Langmuir model was the best-fit isotherm model for E120 dye adsorption into activated carbon which indicated that all the adsorption sites are equivalent and therefore homogenous. The R² values for the Freundlich and Temkin isotherm-fits indicated that these models are less suitable for the current system. The values of n obtained for the Freundlich model to the E120 dye adsorption process over the tested concentration range (10 to 80 mg/L) showed that the surface of activated carbon was reasonably homogeneous, in 293 - 303 K as far as the dye molecules were concerned [16]. The values of Temkin isotherm model A and B indicated that the heat of sorption results from physical interaction between the activated carbon and E120 dye (Table 1).

Thermodynamic parameters of carmine dye adsorption

The values of enthalpy ΔH (KJ/mol) and entropy ΔS (J/mol.K) for the adsorption of E120 dye into activated carbon were calculated as in Eq 3.

$$\ln K_d = \Delta S^\circ/R - \Delta H^\circ/RT \dots\dots (3)$$

R represents the gas constant (8.314 J/mol.K), T is the absolute temperature and is measured by (Kelvin), while K_d is the distribution coefficient. ΔH° and ΔS° can be calculated from the slope and intercept of the plot of ln K_d versus 1/T, respectively. As shown in Table 2, the values of ΔH° (kJ/mol) and ΔS° (J/mol.K) were 8.7 and -27.8, respectively. The positive value of ΔH° indicated that the E120 dye adsorption onto active carbons was endothermic. The energy of dehydration (liberation of the previously adsorbed water) is higher than the energy of adsorption (bond formation between E120 dye and surface of activated carbons)[9]. The enthalpy of adsorption of organic molecules from aqueous solution on activated carbon range from 8 – 65 kJ/mol [10]. The negative sign of entropy indicated the decreased randomness at the solid–liquid interface during E120 dye adsorption

on activated carbon. The decreased randomness was due to increased liberation of the previously adsorbed water molecules upon dye adsorption [17]. The positive sign of free energy values (ΔG) indicated that the adsorption process is not spontaneous. The free energy values (ΔG) increased from 16.86 KJ/mol at 293 K to 17.14 KJ/mol at 303 K again indicating that the adsorption of E120 dye onto activated carbon was endothermic.

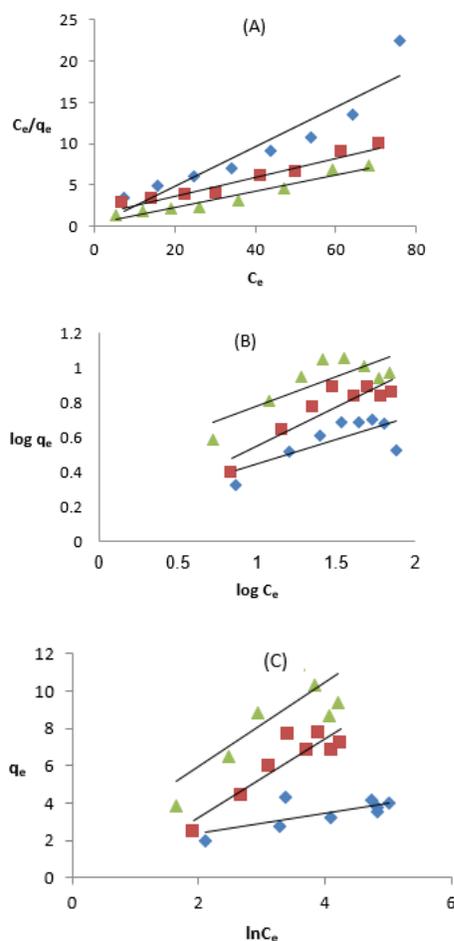


Figure 8: Langmuir (A), Freundlich (B), and Temkin (C) models of carmine dye adsorption onto activated carbon at 293k (♦), 298k (■), and 303K (▲)

Table 1: Langmuir, Freundlich and Temkin parameters

T(K)	Langmuir			Freundlich				Temkin			
	R ²	q _m (mg/g)	K _L (L/mg)	T(K)	R ²	K _f (mg/g)	n	T(K)	R ²	A (L/g)	B (J/mol)
293	0.8705	4.18	2.30	293	0.5656	0.1574	3.48	293	0.5165	2.08	0.5462
298	0.9563	8.73	0.08	298	0.8194	0.1026	2.23	298	0.8216	0.33	2.1203
303	0.9491	10.11	0.31	303	0.6592	0.4453	2.98	303	0.5419	4.59	2.2321

Adsorption kinetic models

Kinetics models were investigated to evaluate the adsorption of E120 dye into activated carbon. The pseudo-first order and pseudo-second order were used to evaluate the experimental data.

Pseudo first-order

Values of k_1 and q_e (mg/g) were calculated by plotting a graph of $\log (q_e - q_t)$ versus 't' (hours) as in Eq 4.

$$\log [q_e - q_t] = \log [q_e] - [k_1/2.303]t \dots\dots (4)$$

where q_e and q_t are the adsorption capacity at equilibrium and at time t, respectively (mg/g). k_1 is the pseudo first-order rate constant. The rate constants (k_1) and q_e can be calculated from the slope and intercept of the linear plot, respectively. As shown in Table 3, the value of experimental q_e' value (mg/g) does not agree with the calculated ' q_e ' (mg/g) plot. Also, Figure 9 showed that $R^2 = 0.9946$ indicates that the first order Kinetic model does not fit with the adsorption of E120 dye into activated carbon.

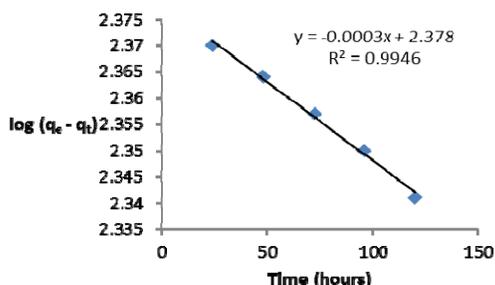


Figure 9: Pseudo-first- order kinetics for E120 dye adsorption onto activated carbons

Pseudo-second-order kinetic model

Pseudo-second-order kinetics must be studied if the adsorption involves chemical interactions/bonding between the adsorbent and adsorbate (Eq 5).

$$t/q_t = 1/k_2 q_e^2 + 1/q_e t \dots\dots (5)$$

where q_e , q_t , and t, were explained above, k_2 is the rate constant of pseudo second-order kinetic

adsorption [g/mg.min]. Values of k_2 and q_e (mg/g) were calculated by plotting t/q_e versus 't' (hours) which will give a linear plot if the model is valid (Figure 10). The rate constants (k_2) and q_e can be calculated from the slope and intercept of the linear plot, respectively, (Table 2). The value of experimental q_e' (mg/g) was in agreement with the calculated ' q_e ' (mg/g). Figure 10 showed that $R^2 = 0.9885$ indicating that pseudo-second order kinetic model is the suitable kinetic model for E120 into activated carbon.

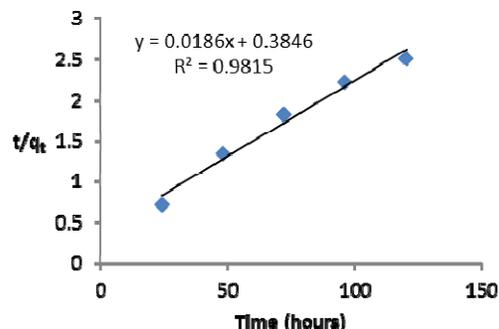


Figure 10: Pseudo second- order kinetics for E120 dye adsorption onto activated carbons

Comparison of carmine dye adsorption with various adsorbents

The adsorptive capacities of the adsorbents used in this work have been compared with those of others reported in the literature and the values of adsorption capacity are presented in Table 4. The adsorption capacity (q_m) is used as comparative parameter. The q_m value obtained in this study agrees with most previous studies but higher than most of them, which indicated that the activated carbon used in this study easily removed the E120 dye from solutions.

DISCUSSION

Adsorption isotherms of the E120 dye on activated carbon was better fitted of the equilibrium adsorption data to the Langmuir isotherm model than the Freundlich and Temkin isotherm models and gave a maximum adsorption capacity of 4.18 mg/g at 293 K which increased to 10.11 mg/g at 303 K and pH = 7.

Table 3: Parameters for various kinetic models

Initial dye conc. (mg/L)	Pseudo-First-order			Pseudo-Second-order		
	k_1 (min^{-1})	Calculated q_e (mg/g)	R^2	k_2 (g/mg. min)	Calculated q_e (mg/g)	R^2
80	5.48	0.99	0.9946	8.99×10^{-4}	53.76	0.9815

Table 4: Comparison of adsorption capacity of different adsorbents for the adsorption of carmine dye

Adsorbent	Adsorption capacity q_m (mg/g)	Reference
Fly Ash (FA)	1.48	[18]
Synthesized zeolite (ZM)	1.23	[18]
Apricot Stone Active Carbon	552.5	[4]
Cola nut shells (treated with H_3PO_4)	9.997	[4]
Active carbon (treated with H_3PO_4)	5.861	[4]
Active carbon (treated with KOH)	12.093	[4]
Calcium hydroxide	4.865	[12]
Activated Carbon	10.11	This work

Adsorption of E120 dye onto activated carbon was non-spontaneous (positive ΔG) and endothermic (positive ΔH) in nature at the examined temperature range.

The negative entropy (ΔS) state clearly that the decreased randomness at the solid-solution interface during E120 dye adsorption onto the activated carbon. Adsorption of E120 dye onto activated carbon obeyed pseudo second-order kinetic model.

CONCLUSION

This study demonstrates the applicability of activated carbon as a sustainable adsorbent for the removal of E120 dye from aqueous solution. The adsorption could be described by Langmuir, Freundlich and Tempkin isotherm with a maximum adsorption capacity of 4.18 mg/g.

The thermodynamic investigation showed that adsorption of E120 dye onto activated carbon was non-spontaneous (positive ΔG) and endothermic (positive ΔH) in nature at the examined temperature range.

DECLARATIONS

Acknowledgement

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Conflict of interest

No conflict of interest is associated with this work.

Contribution of authors

We declare that this work was done by the authors named in this article and all liabilities pertaining to claims relating to the content of this article will be borne by the authors. Mater H Mahnashi: Conceptualization, Project administration, Writing - original draft, Writing - review & editing, Formal analysis, Visualization, Validation. Samer S Abu-Alrub: and Mohammad W Amer conceived this work, did the laboratory work and data analysis as well as the manuscript writing which was read and approved by all authors for publication. Ali O Alqarni was involved in data analysis, writing of the manuscript and final review of the manuscript before submission for publication.

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