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Research Article

Adsorption of food coloring allura red dye (E129) from aqueous solutions using activated carbon

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Abstract

The adsorption behavior of Allura red (E129) from aqueous solutions onto activated carbon was successfully investigated. All factors affecting the adsorption process were carefully studied and the conditions were optimized. Adsorption of E129 onto activated carbon was found to increase by decreasing the mass of activated carbon, pH and ionic strength of the solution and by increasing temperature. Under the optimum conditions, the maximum adsorption capacity of activated carbon for E129 dye was 72.85 mg.g^{-1}. Three adsorption isotherm models; Langmuir, Freundlich and Temkin model were investigated regarding E129 adsorption onto activated carbon. The models' parameters K_L, qm, R2, and (n) were determined and found to be 0.0222, 72.85 mg.g^{-1}, 0.9057-0.9984, and 0.992, respectively. Moreover, pseudo first-order and pseudo second-order kinetic models were applied to the adsorption data. The obtained data from adsorption isotherms, kinetic models, and thermodynamic parameters (the free energy change, enthalpy and entropy) indicated that E129 adsorption is spontaneous exothermic heterogeneous adsorption. Furthermore, the results showed that E129 adsorption onto activated carbon obeyed both Freundlich isotherm and pseudo second-order kinetic models.

1. Introduction

Allura red is a synthetic water-soluble food dye has the E-number: E129. E129 dye was originally introduced in the USA as an alternative for amaranth E123 as a food coloring agent used in food and condiments, medications and cosmetics. E129 dye has other names such as food red 17, **C.I.** 16035, and FD&C Red 40 (**Figure** 1). (Food Color Facts, 2007). It is widely used in bakery products, dairy products, meat and fish products, soft drinks, jams, jellies, chewing gum and canned food (Al-Degs, 2009; Bernstein et al., 1975).

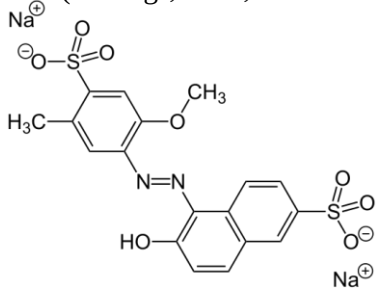

Figure. 1 The chemical structure of Allura red (E129) dye

E 129 dye is toxic to human health, if it is excessively consumed (Al-Degs, 2009), and may be a cause for cancer (Amate et al., 2010). Therefore, safety data sheet of E129 including the acceptable daily intake amount based on toxicological data have been determined and evaluated by Food Agricultural Organization (FAD) and World Health Organization (WHO) (Al-Degs, 2009). Moreover, E 129 dye can cause human sensitivity to light and affect thyroid hormones resulting in hyperthyroidism (European Parliament and Council, 1994). The European Union is opposed to the use of E129 in meat products and processed food (Butt et al., 2005).

Owing to the growing health hazards of the excessively consumed dyes, disposal of dyes in precious water resources must be avoided. Therefore, various technologies were used for the dye uptake from aqueous waste solutions such as biological, physical, chemical, and radiation treatments (Gupta and Suhas, 2009). These technologies include: coagulation/ flocculation (Wu et al., 2008), cation or anion exchange (Fan et al.,

2008), electrochemical degradation (Banerjee et al., 2007), oxidation, ozonization, filtration (Mahmoud et al., 2007; Fathima et al., 2008; Lodha & Chaudhari, 2007), Fenton-biological treatment (Garcia-Montano et al., 2008; Allen et al., 2004), and adsorption (Erdem et al., 2005; Hameed, 2009a; Hameed et al., 2009; Alzaydien, 2009). Among various methodologies, adsorption has been becoming of great interest in removing of dyes from industrial wastewater. Adsorption is considered one of the best methods used for dye uptake from aqueous solutions (Alzaydien, 2009; Malik, 2004) as it takes into account both economic and environmental concerns. Several adsorbents have been used for dye removal from aqueous solutions such as silica gel, alumina, zeolite, chitosan, a natural aminopolysaccharide, gelatin microspheres, polyamide solid phase extraction (SPE) cartridges, and activated carbons (Gupta and Suhas, 2009; Crini and Badot, 2008; Dotto and Pinto, 2011; Xue et al, 2012; Zalacain et al, 2005; Issa & Al-Degs, 2009). Activated carbons is the oldest microporous adsorbent used not only in removing of dyes, but also other organic pollutants such as pesticides, chlorinated hydrocarbons, detergents (Gupta and Suhas, 2009). Adsorption on activated carbons has been reported as an efficient methodology for dye removal from aqueous solution depending on different conditions, such as concentration, pH, ionic strength, and temperature of the solution (Issa & Al-Degs, 2009). The adsorption of E129 dye was previously studied using different adsorbents such as: SPE Amberlite XAD resins (Bişgin et al., 2015), polyamide SPE cartridges (Zalacain et al, 2005), hanging dropping mercury electrode (Alghamdi, 2005), macroporous polystyrene anion exchangers Amberlite IRA (Wawrzkiewicz, & Hubicki, 2009) and chitosan (Liang et al., 2015). Yet, the adsorption of E129 dye onto activated carbons has not been studied. Therefore, the present work was devoted to explore the use of low-cost activated carbon adsorbent in adsorption and removal of E129 dye from aqueous solutions. Isotherms models, thermodynamic parameters, and kinetic models of the adsorption process were studied.

2. Materials and methods

2.1. Chemicals and reagents

All chemicals and solvents used in this study were of analytical grade. Allura red (E129) dye was obtained from Lobal Chemie (India). Activated carbon with particle diameter = 300 - 500µm was purchased from Nen Tech Ltd (UK).

2.2. Apparatus

A double beam UV-vis spectrophotometer (SP-300, Optima, Japan) was used for obtaining absorption spectrum of E129. pH measurements were carried out with a Lovibond pH meter (Germany). A wise Bath shaker (Daihan Scientific, Korea) was used in this study.

2.3. Determination of adsorption of E129 dye

To determine the concentration of E129 dye in aqueous solutions, six concentration levels (three replicates for each) of E129 dye were used (1.0-100.0 mg L^{-1}) to construct the calibration curve. The calibration curve was obtained by plotting the absorbance of the dye solutions at 505 nm (λ_{max} of E129 dye) versus the corresponding dye concentrations. Beer's law is obeyed with a good correlation coefficient equal 0.9885.

The adsorption of E129 dye was studied using the batch method. Four hundreds milligrams of activated carbon with particle diameter 300-500 µm was mixed with 100 mL of the dye solution and the mixture was agitated for 4 days, at pH 7 and 25ºC. After completion of agitation, the solution was carefully removed and filtered. The remaining dye concentration (equilibrium concentration) in the clear solution was determined spectrophotometrically at 505 nm using the linear regression equation resulted from the abovementioned calibration curve. The amount of adsorbed dye (Qe, $mg.g^{-1}$) could be calculated by difference between the initial (C0, $mg.L^{-1}$) and equilibrium (Ce, $mg.L^{-1}$) dye concentration. The adsorption of E129 dye was expressed in term of the distribution coefficient K_d ($L.g^{-1}$).

Amount of adsorbed dye (Q_e) = (C_o − Ce) * V (L) / mass of adsorbent (g)

Distribution coefficient (K_d) = Qe / Ce

2.4. Effect of contact time on E129 dye uptake

Hundred mL of the dye solution (300 $mg.L^{-1}$) was agitated for different times (1-7 days) with. 0.4 g of the activated carbon with particle diameter 300-500 µm at pH 7 and 25ºC. The remained dye concentration was determined at 505 nm.

2.5. Effect of the mass of activated carbon on E129 dye uptake

Different masses (0.20-0.90 g) of the activated carbon with particle diameter 300-500 µm were mixed with 100 mL of the dye solution (300 $mg.L^{-1}$). The mixtures were agitated for 4 days, at pH 7 and 25ºC. The remained dye concentration was determined at 505 nm.

2.6. Determination of adsorption isotherms

Four hundreds milligrams of activated carbon with particle diameter 300-500 µm was added to set of dye solutions (50-450 $mg.L^{-1}$). The mixtures were agitated for 4 days, at pH 7 and 25ºC in a final solution volume of 100 mL. The remained dye concentration was determined at 505 nm

2.7. Effect of ionic strength on E129 dye uptake

Different concentrations of sodium chloride (0.5-3.0 $mol.L^{-1}$) were mixed with the dye solution in a final volume of 100 mL. The solutions were agitated with 0.4 g of activated carbon of particle diameter 300 – 500µm for 4 days at 25ºC and pH 7. The remained dye concentration was determined at 505 nm

2.8. Effect of pH on the dye uptake

Series of 100 mL solution of E129 dye (300 $mg.L^{-1}$) were prepared at different pH values (1, 3, 7, 9, and 11) using buffer solutions. The solutions were agitated with 0.4 g of activated carbon of particle diameter 300 – 500µm for 4 days at 25ºC. The remained dye concentration was determined at 505 nm

2.8. Effect of temperature on E129 dye uptake

Series of 100 mL solution of E129 dye (300 $mg.L^{-1}$) were prepared at pH 7.0. The solutions were agitated with 0.4 g of activated carbon of particle diameter 300 – 500µm for 4 days at different temperatures (20, 30, and 40 ºC). The remained dye concentration was determined at 505 nm.

3. Results and discussion

3.1. Adsorption properties of E129 dye and activated carbon

Al-Degs reported that commercial activated carbon has both basic and acidic functional groups. The basic groups are more abundant. The large surface area and availability of many surface functional groups makes the activated carbon a potential adsorbent for toxic dyes and able to adsorb E129 dye (Al-Degs et al., 2008). Allura red dye is a commercial foodstuffs dye containing an azo-group accounting for its coloring properties. Therefore, it is considered an electrochemical active compound with irreversible reaction (Zhang et al., 2010). These properties makes the Allura red highly toxic compounds with possible carcinogenic effect (Amate et al., 2010). Therefore, the removal of the Allura red dye from aqueous solutions has a great importance. E 129 dye adsorption onto different adsorbents was reported using different analytical techniques such as potentiodynamic polarization (Peme et al., 2015), spectrophotometry (Bişgin et al., 2015, Zalacain et al., 2005), electrochemical methods (Alghamdi, 2005), HPLC (Zhang, 2015) and Fourier transform-infrared spectroscopic (FT-IR) analysis (Liang et al., 2015). Among these techniques, spectrophotometry stands out, as the most effective and convenient analytical techniques as compared to the others owing to its simplicity, low cost, and wide availability in most laboratories. The efficiency of the adsorbents is considered acceptable when the distribution coefficient (K_d) value is higher than unity with minor exceptions. Therefore, the aim of the present investigation was devoted to explore the use of low cost adsorbent as activated carbon in the removal of E129 from aqueous solutions.

3.2. Optimization of the conditions

3.2.1. Effect of contact time on the dye uptake

To determine the enough agitation contact time between E129 dye and activated carbon required for the maximum dye uptake, different times (1-7 days) were investigated. The results shown in **Figure 2** revealed that the adsorption of E129 increased by increasing the contact time until become nearly constant at times ≥ 4 days. This indicated that the adsorption of E129 dye on the activated carbon reached equilibrium after 4 days.

3.2.2. Effect of mass of activated carbon on the dye uptake

To determine the mass of activated carbons required for the maximum dye removal, different masses (0.20-0.90 g) of the activated carbon were tested. **Figure 3**

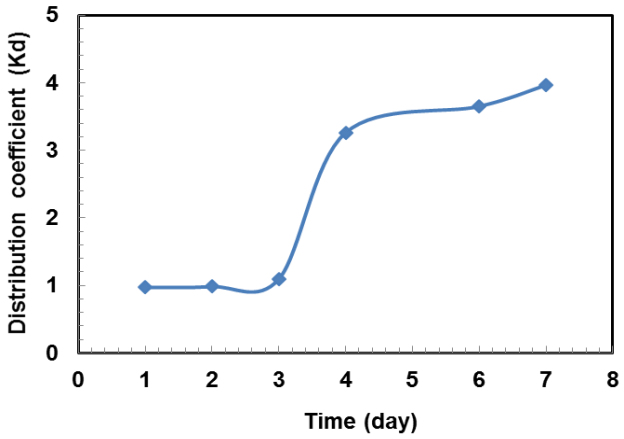

Figure 2 Effect of contact time on E 129 adsorption onto activated carbon

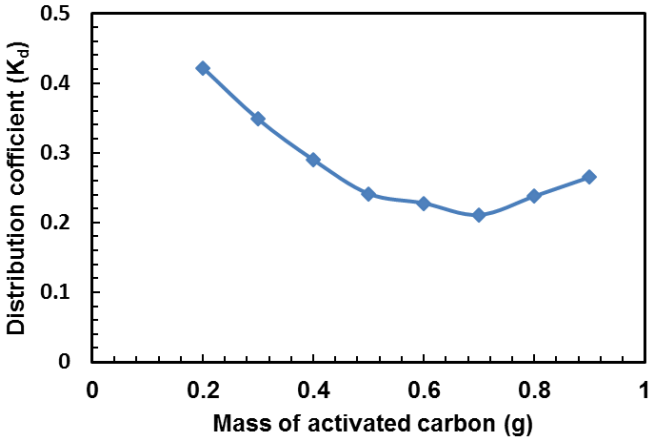

Figure 3. Effect of mass of activated carbon on E129 adsorption

indicated that the dye uptake expressed as distribution coefficient decreased markedly with increasing the mass of the activated carbons. Accordingly, further experiments were carried out using 0.4 g of the activated carbon as the optimum mass.

3.2.3. Effect of dye concentration on the dye uptake (adsorption isotherm)

To evaluate the adsorption isotherm, different initial concentrations of E129 dye (50-450 $mg.L^{-1}$) were studied while keeping the other variables constant. The distribution coefficient (Kd) values were calculated at each concentration. **Figure 4** showed that the distribution coefficient increased significantly as the dye concentration increased up to 300 $mg.L^{-1}$, thereafter it becomes constant. Therefore, further experiments were carried out using dye solution concentration of 300 $mg.L^{-1}$.

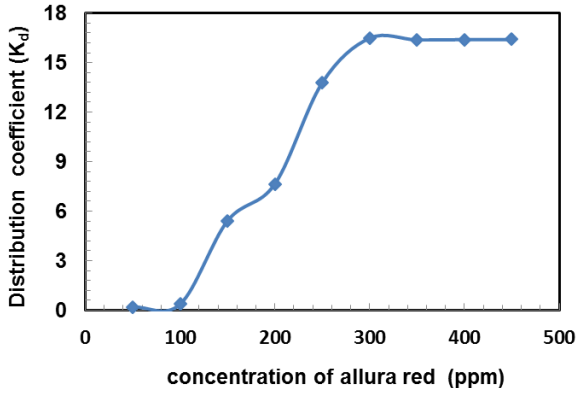

Figure 4. Effect of E129 concentration on the dye uptake onto activated carbon

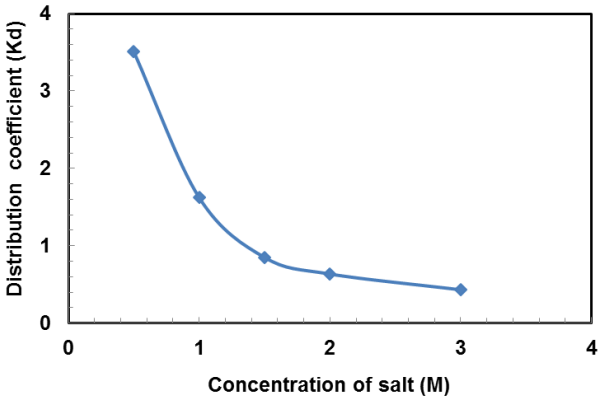

Figure 5 Effect of ionic strength of the aqueous solution on E129 adsorption onto activated carbon

3.2.4. Effect of ionic strength on E129 dye uptake

To investigate effect of ionic strength on E129 dye uptake, different ionic strengths of sodium chloride solutions were tested. **Figure 5** showed that the adsorption on the activated carbon decreased when the ionic strength of sodium chloride was more than 0.5M in the mixture. This may be due to the increased attractive forces between molecules resulting in increased inhibition of adsorption at the carbon surface (Fuqiang et al., 2004).

3.2.5. Effect of pH on E129 adsorption by activated carbon

The results indicated that the adsorption of E129 by activated carbon is pH dependent as shown in **Figure 6**. The increased E129 adsorption on the activated carbons in acidic medium may be owing to the electrostatic interactions between the protonated surface of the activated carbon and the negatively charged E129 molecules, enhancing the adsorption on the activated carbon. However, the decreased E129 adsorption in

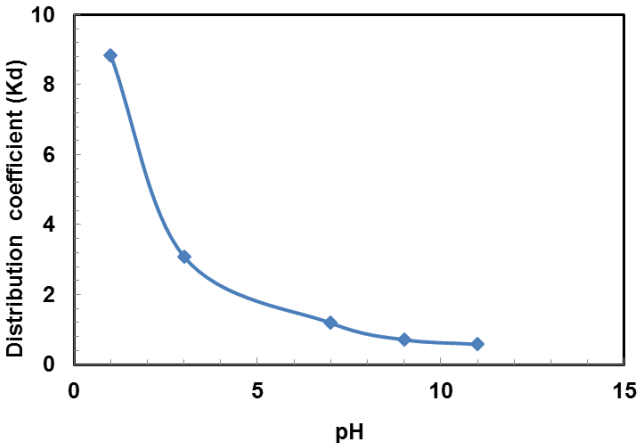

Figure 6. Effect of pH on E129 adsorption onto activated carbon

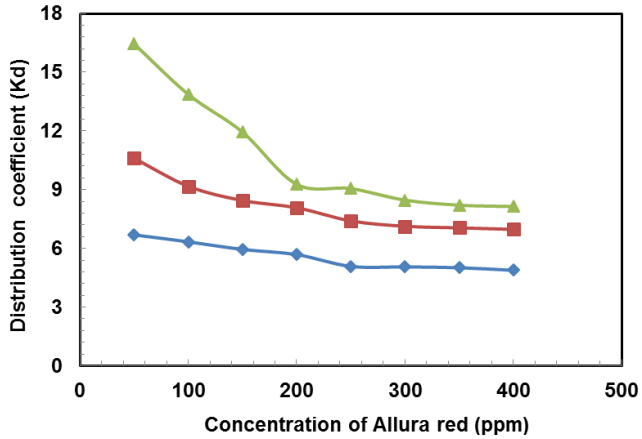

Figure 7. Effect of temperature on E129 adsorption onto activated carbon at 20 °C (-♦-), 30 °C (-■-), and 40 °C (-▲-).

alkaline medium may be due to the electrostatic repulsion between the deprotonated E129 molecule and negatively charged activated carbon because of adsorption of hydroxyl ions on the surface (Fuqiang et al., 2004).

3.3. Adsorption isotherms

It is well known that the adsorption process, expressed in distribution coefficient, is highly influenced by temperature. In addition, investigating the effect of temperature on the adsorption may determine if the process is physical or chemical adsorption. Therefore, the adsorption isotherm was studied for E129 in a concentration ranged from 50 to 450 $mg.L^{-1}$ at different temperatures (20 ºC, 30 ºC, 40 ºC). Adsorption data presented in **Figure** 7 showed that the distribution coefficient of E129 increased by increasing temperature. In addition, the results revealed that the temperature had a great effect on the distribution coefficient of E129 at low concentrations and a lower effect at high concentrations. This because higher temperatures may enhance the penetration of E129 inside micro pores of the adsorbent or promote creation of new active sites on the active carbon. This adsorption behavior proved that the adsorption of E129 onto activated carbons is not a physical adsorption, but a chemical interaction must be existing between activated carbon and E129 molecule.

3.3.1. Evaluation of adsorption isotherms

Assessment of the favorability of the adsorption process depends mainly on the distribution coefficient (K_d) value of the adsorption isotherm. If $K_d > 1$, this means that E129 dye favors adsorption onto active carbon surface over solution. The adsorption data were modeled using Langmuir (assuming similar adsorption sites), Freundlich (assuming varying adsorption sites) and Temkin (assuming adsorbent-adsorbate interaction) isotherms to determine the maximum adsorption capacity of activated carbon with respect to E129.

Langmuir model: Use the equation:

$$K_d = q_e/C_e = q_m K_L - K_L q_e \qquad (1)$$

Where K_d is the distribution coefficient, q_e is the amount of E129 adsorbed per gram of activated carbon at equilibrium, C_e is the concentration of E129 in the solution at equilibrium, q_m is the maximum adsorption capacity corresponding to complete saturation of the activated carbon surface, K_L is the Langmuir equilibrium constant. This model was evaluated by plotting of (K_d) against (q_e) to obtain a straight line with slope = - K_L and an intercept = $q_m K_L$ (**Figure 8**).

Freundlich model: Use the equation

$$\log q_e = \log K_F + 1/n \log \qquad (2).$$

This model was evaluated by plotting of (log q_e) versus (log C_e) to obtain a straight line as shown in **Figure 8** with a slope 1/n and intercept (log K_F); where K_F and n are the Freundlich constants. Affinity of E129 dye towards the activated carbon was expressed by the value of n (Özbay et al., 2013).

Temkin model: Use the equation

$$q_e = B \ln A + B \ln C_e \qquad (3)$$

This model was evaluated by plotting of (q_e) versus (ln C_e) to obtain a straight line as shown in **Figure 8** with a slope (B) and intercept (B ln A); where A is Temkin constant used to examine the interaction between active carbon and E129 dye, and (B) is a constant related to adsorption heat.

Evaluation of the employed models was mainly based on the value of the correlation coefficient. The highest correlation coefficient value indicate the best-fit isotherm model for adsorption of E129 dye. Comparing R^2 values of the evaluated models indicated that Freundlich model was the best fit isotherm model for E129 dye adsorption onto activated carbon than both Langmuir and Temkin models, where R^2 values of Freundlich isotherm were the highest (0.9786 – 0.9984) among all evaluated isotherms (Table 1). The adsorption parameters; q_m, K_L, n, K_f, A and B of adsorption isotherms, under the optimum experimental conditions, were also calculated and tabulated in Table 1. Depending on this conclusion, we can say that the adsorption of E129 dye onto activated carbons is a heterogeneous adsorption. This means that the interaction between activated carbons and E129 molecules is varied from adsorbing site to another. This was previously confirmed for the activated carbon (Özbay et al., 2013). This heterogeneous Freundlich adsorption behavior of E129 onto activated carbon was also confirmed by the obtained n-value that was less than unity due to the high variation in the energies of the active sites on the surface of activated carbons (Yang & Al-Duri, 2005; Haghseresht & Lu, 1998). The low *n*- value revealed that the adsorption of E129 is favorable over the entire concentration range used in this study (McClellan & Harnsberger, 1967).

3.3.2. Thermodynamic parameters of E129 adsorption

It is expected that the adsorption of E129 onto activated carbons include bond formation process. Consequently, it is expected that the adsorption of E129 will be associated with a change in the free energy (ΔG), enthalpy (ΔH°) and entropy (ΔS°) of the adsorption system. Therefore, it was important to determine the thermodynamic parameters (ΔG, ΔH°, and ΔS°) of E129 adsorption onto activated carbons. Under the optimum conditions, enthalpy and entropy were calculated using distribution coefficient (K_d) in the following equation (4):

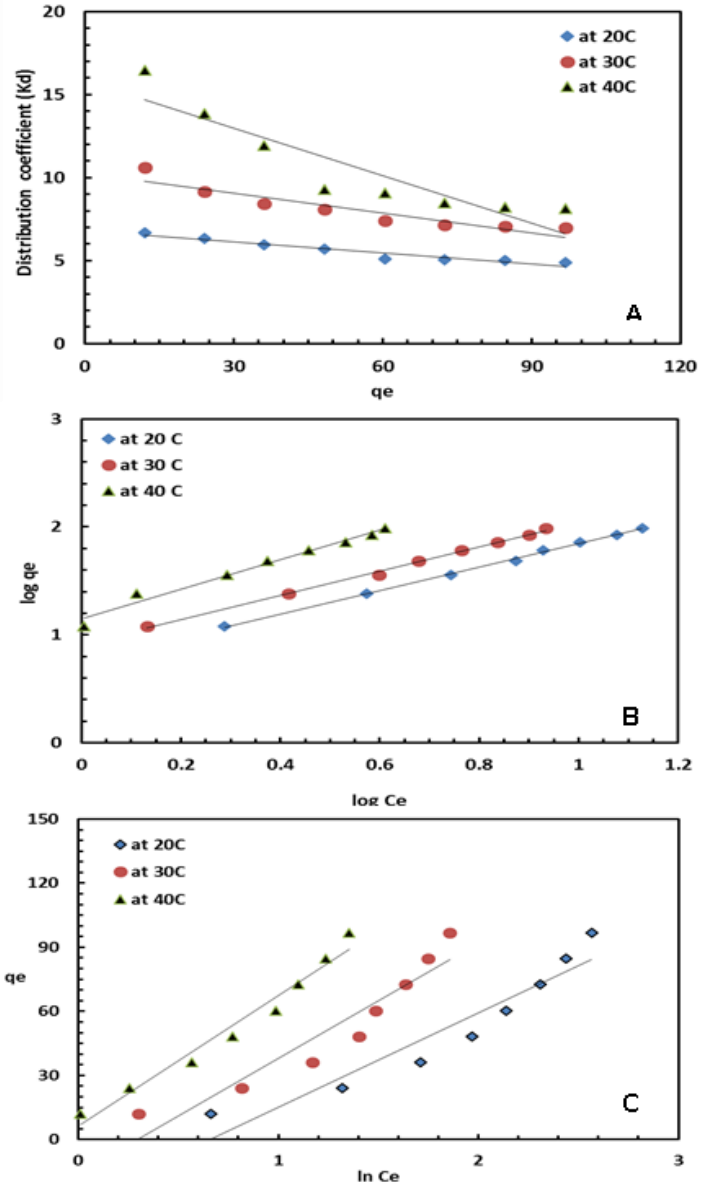

Figure 8 Langmuir (A), Freundlich (B), and Temkin (C) models of E129 adsorption onto activated carbon at 20 °C (♦), 30 °C (●), and 40 °C (▲).

$$\ln K_d = \Delta S^\circ/R - \Delta H^\circ/RT \qquad (4)$$

Where R is the gas constant (8.314 $J.mol^{-1}$ Kelvin), T is absolute temperature (Kelvin) and K is the equilibrium constant. ΔH° and ΔS° can be calculated from the slope and intercept of the plot of ln K_d versus 1/T, respectively. Figure 9 shows the distribution coefficient - temperature plot of the E129 adsorption onto activated carbons under the optimized conditions. The obtained values of ΔH° ($KJ.mol^{-1}$) and ΔS° ($J.mol^{-1}.K^{-1}$) were – 2.37 and 9.74, respectively. The negative sign of ΔH° indicated that the E129 adsorption onto active carbons was exothermic process, i.e. the energy of adsorption (bond formation between E129 and surface of activated

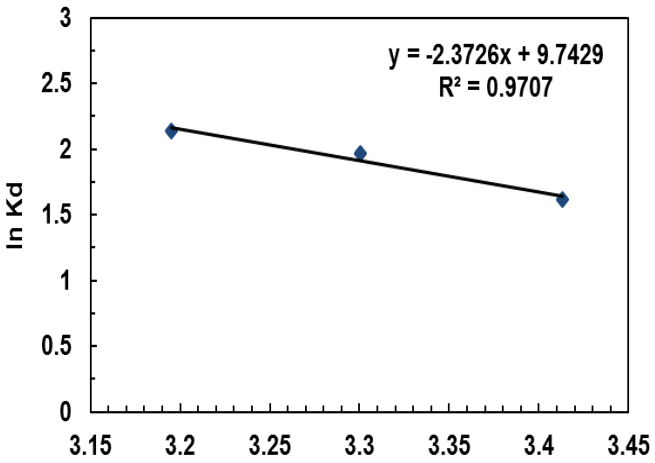

Figure 9 Distribution coefficient-temperature plot of E129 adsorption onto activated carbons under the optimized conditions

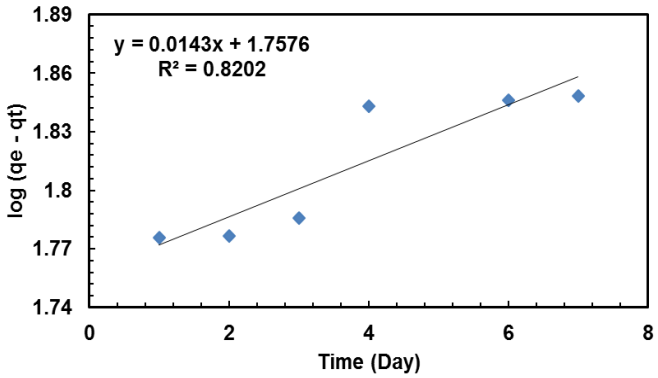

Figure 10 Pseudo first-order kinetic plot for E129 adsorption onto activated carbons

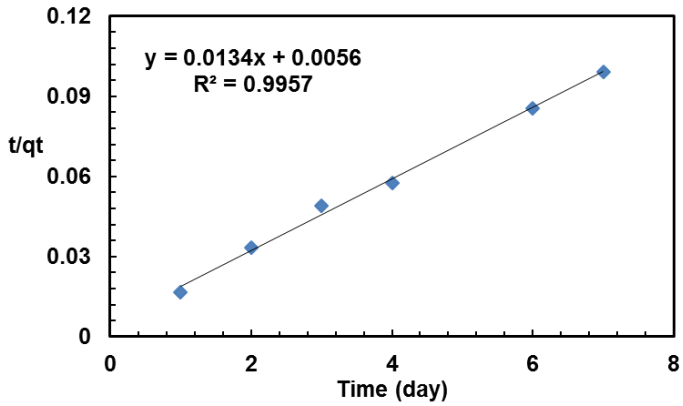

Figure 11 Pseudo second-order kinetic plot for adsorption of E129 onto activated carbons

carbons) is higher than the energy of dehydration (liberation of the previously adsorbed water). However, the positive value of entropy of E129 adsorption is an indicative measure for the increased randomness at the solid–liquid interface during E129 adsorption. The increased randomness may be owing to structural changes in both E129 and activated carbons during the adsorption process.

The free energy change (ΔG°) can be calculated using the following equation (5)

$$\Delta G^{\circ} = \Delta H^{\circ} - T\,\Delta S^{\circ} \qquad (5)$$

The values of ΔG ($KJ.mol^{-1}$) was (-5.32 – 5.42). The free energy change for E129 adsorption decreased by increasing the temperature indicating thermodynamically favorable and spontaneous adsorption process.

3.4. Adsorption kinetic models

To study the mechanism and the rate by which the adsorption of E129 dye onto activated carbons occurred (mass transfer, diffusion control, or chemical reaction), different kinetic models; pseudo first-order and pseudo second-order kinetic reaction were applied to adsorption data (McClellan & Harnsberger, 1967).

3.4.1. Pseudo first-order kinetic model

To study pseudo-first order kinetic model, the following equation is generally used:

$$\text{Log}\,[qe - qt] = \log\,[qe] - [k1/2.303]t \qquad (6)$$

Where qe and qt are the adsorption capacity at equilibrium and at time t, respectively ($mg.g^{-1}$). k_1 is the pseudo first-order rate constant. Plotting of log (qe-qt) versus t should give a linear plot, if the adsorption process obeyed pseudo first-order kinetic model. The rate constants (k_1) and qe can be calculated from the slope and intercept of the linear plot, respectively. Table 2 shows the calculated Kinetic parameters of the applied pseudo first-order kinetic model and Figure 10 showed that E129 adsorption onto activated carbons did not obey pseudo first-order kinetic model.

3.4.2. Pseudo second-order kinetic model

To investigate pseudo second-order kinetic model, the following linear equation is used:

$$t/qt = 1/k2\ qe2 + 1/qe\ t \qquad (7)$$

Where qe, qt, and t, were explained above, k_2 is the rate constants of pseudo second-order kinetic adsorption

Table 1. Langmuir, Freundlich and Temkin adsorption parameters for E129 dye adsorption**.**

T (° C)	Langmuir isotherm		Freundlich isotherm			Temkin isotherm		
	R^2	K_L	R^2	K_f	*n*	R^2	A	B
20	0.9224	0.0222	0.9984	0.7602	0.922	0.9057	1.93	44.114
30	0.8672	0.0392	0.9976	0.9219	0.89	0.8591	1.41	55.615
40	0.8365	0.0933	0.9786	1.1593	0.73	0.9701	1.1	62.666

Table 2. The calculated Kinetic parameters of E129 adsorption onto the activated carbons

Initial E129 Conc. ($mg.L^{-1}$)	Experimental qe ($mg.g^{-1}$)	Pseudo first-order kinetic model			Pseudo second-order kinetic model		
		k_1	Calculated qe ($mg.g^{-1}$)	R^2	k_2	Calculated qe ($mg.g^{-1}$)	R^2
300	12.25	0.0329	1.7576	0.8202	$1.01X10^{-6}$	74.62	0.9957

[$g.mg^{-1}.min^{-1}$]. Plotting of t/qt versus t resulted in a linear plot as shown in Figure 11. The pseudo second-order rate constant and qe can be determined from the intercept and slope of the plot, respectively (Table 2). Evaluation of the applied kinetic models was mainly based on the value of the regression coefficient and the calculated maximum adsorption capacity compared to the experimental one. The kinetic adsorption model with the highest regression coefficient indicates the best-fit kinetic model for E129 dye adsorption. Comparing R^2 values of the evaluated kinetic models indicated that the E129 adsorption onto activated carbon obeyed pseudo second-order kinetic model (Table 2 and Figures 10 and 11).

4. Conclusions

E129 dye adsorption from aqueous solutions onto activated carbon was found to be acceptable with maximum adsorption capacity of 72.85 $mg.g^{-1}$. Adsorption of E129 increases by decreasing pH of the solution and by increasing temperature. The temperature had a great effect on E129 adsorption at low concentrations and a lower effect at high concentrations of E129. Adsorption of E129 is an exothermic heterogeneous adsorption process, where the energy of adsorption is higher than the energy of dehydration. The adsorption of E129 dye is mainly attributed to its large molecular structure and its chemical interaction with active sites on activated carbon. Adsorption of E129 onto activated carbon obeyed both Freundlich isotherm and pseudo second-order kinetic models.

5. Acknowledgments

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7. References

Al-Degs, Y. (2009). Determination of three dyes in commercial soft drinks using HLA/GO and liquid chromatography *Food Chem. Anal Meth*, 117: 485–490.

Al-Degs, Y. S., El-Barghouthi, MI., El-Sheikh, AH. & Walker, G. (2008). Effect of solution PH, ionic strength, and temperature on adsorption behavior of reactive dyes on activated carbon. *Dyes Pigm*., 77: 16-23.

Alghamdi, A. H. (2005) Determination of allura red in some food samples by adsorptive stripping voltammetry. *J AOAC Int*. 88: 1387-93

Allen, S. J., Mckay, G. & Porter, J. F. (2004). Adsorption isotherm models for basic dye adsorption by peat in single and binary component systems. *Journal of Colloid and Interface Science*, 280: 322–333.

Alzaydien, A.S. (2009). Adsorption of methylene blue from aqueous solution onto a low-cost natural Jordanian Tripoli. *Am. J. Environ. Sci*., 3: 197-208.

Amate, C. F., Unterluggauer, H., Fisher, R. J., Fernandez-Alba, A. R. & Msselter, S. (2010). Development and validation of a LC-MS/MS method for the stimulants determination aflatoxins, dyes and pesticides in spices. *Anal. Bioanal. Chem*., 397: 93-107.

Banerjee, P., Dasgupta, S. & De, S. (2007). Removal of dye from aqueous solution using a combination of advanced oxidation process and nanofiltration. *Journal of Hazardous Materials*, 140: 95-103.

Bernstein, R., Haugen, H. & Frey, H. (1975). Clinical Chemistry: Thyroid Function during Erythrosine Ingestion in Doses Encountered in Therapy with Conventional Antibiotics. *Scan. J. Clini. Lab. Invest*., 35: 49-52.

Bişgin, A.T., Uçan, M. & Narin, İ. (2015) Comparison of Column Solid-Phase Extraction Procedures for Spectrophotometric Determination of E129 (Allura Red) in Foodstuff, Pharmaceutical, and Energy Drink Samples. *J AOAC Int*., 98: 946-952

Butt, M. T., Arif, F., Shafique, T. & Imtiaz, N. (2005). Spectrophotometric estimation of colour in textile dyeing wastewater. *Journal of the Chemical Society of Pakistan*, 27(6): 627-630.

Crini, G. & Badot, P. M. (2008) Application of chitosan, a natural aminopolysaccharide, for dye removal from aqueous solutions by adsorption processes using batch studies: a review of recent literature. *Progress in Polymer Sciences*, 33: 399-447.

Directive 94/36//EC on colours for use in foodstuffs (1994). European Parliament and Council.

Dotto, G. L. & Pinto, L. A. A. (2011) Adsorption of food dyes onto chitosan: Optimization process and kinetic. *Carbohydrate Polymers*, 84: 231-238.

Erdem, E., Çölgeçen, G. & Donat, R. (2005). The removal of textile dyes by diatomite earth. *Journal of Colloid and Interface Science*, 282: 314–319.

Fan, L., Zhou, Y., Yang, W., Chen, G. & Yang, F. (2008). Electrochemical degradation of aqueous solution of Amaranth azo dye on ACF under potentiostatic model. *Dyes Pigments*, 76: 440–446.

Fathima, N. N., Aravindhan, R., Rao, J. R. & Nair, B. U. (2008). Dye house wastewater treatment through advanced oxidation process using Cu-exchanged Y zeolite: A heterogeneous catalytic approach. *Chemosphere*, 70: 1146–1151.

Food Color Facts at the Wayback Machine (archived October 1, 2007).

Fuqiang, L., Jinlong, C., Aimin, L., Zhenghao, F., Junjie, G. & Quanxing, Z. (2004). Equilibrium adsorption of single component and binary mixtures of aromatic compounds onto a polyfunctional Hypercrosslinked polymeric Adsorbent. *Adsorp Sci Tech*., 22: 13-24.

Garcia-Montano, J., Perez-Estrada, L., Oller, I., Maldonado, M. I., Torrades, F. & Peral, J. (2008). Pilot plant scale reactive dyes degradation by solar photo-Fenton and biological processes. *Journal of Photochemistry and Photobiology A: Chemistry*, 195: 205–214.

Gupta, V. K. & Suhas. (2009) Application of low-cost adsorbents for dye removal-A review. *Journal of Environmental Management*, 90: 2313-2342.

Haghseresht, H. & Lu, G. (1998). Adsorption characteristics of phenolic compounds onto coal-rejected-derived adsorbents. *Energy and Fuels*, 12: 1100-1107.

Hameed, B. H. (2009a). Spent tea leaves: A new non-conventional and low-cost adsorbent for removal of basic dye from aqueous solutions. *Journal of Hazardous Materials*, 161:753–759.

Hameed, B. H., Krishni, R. R. & Sata, S. A. (2009). A novel agricultural waste adsorbent for the removal of cationic dye from aqueous solutions. *Journal of Hazardous Materials*, 162: 305–311.

Issa,, A. & Al-Degs, S. (2009). Simple spectrophotometric determination of reactive dyes after preconcentration using activated carbon. *Jordan J Chem*, 4: 89-101.

Liang, T.W., Huang, C.T., Dzung, N.A., Wang, S.L. (2015) Squid pen chitin chitooligomers as food colorants absorbers. *Mar Drugs*. 13: 681-96.

Lodha, B. & Chaudhari, S. (2007). Optimization of Fenton-biological treatment scheme for the treatment of aqueous dye solutions. *Journal of Hazardous Materials*, 148: 459-466.

Mahmoud, A. S., Brooks, M. S. & Ghaly, A. E. (2007). Decolorization of remazol brilliant blue dye effluent by advanced photo oxidation process (H_2O_2/UV system). *American Journal of Applied Sciences*, 4(12): 1054-1062.

Malik, P. K. (2004). Dye removal from wastewater using activated carbon developed from sawdust: adsorption equilibrium and kinetics. *Journal of Hazardous Material*, 113: 81-88.

McClellan, A. & Harnsberger, H. (1967). Cross-sectional areas of molecules adsorbed on solid surfaces. *Journal of Colloid and Interface Science*, 23:577-599.

Özbay, I., Özdemir, U., Özbay, B. & Veli, S. (2013). Kinetic, thermodynamic, and equilibrium studies for adsorption of azo reactive dye onto a novel waste adsorbent: charcoal ash. *Desalination and Water Treatment*, 51: 6091–6100.

Peme, T., Olasunkanmi, L.O., Bahadur, I., Adekunle, A.S., Kabanda, M.M. & Ebenso, E.E. (2015) Adsorption and Corrosion Inhibition Studies of Some Selected Dyes as Corrosion Inhibitors for Mild Steel in Acidic Medium: Gravimetric, Electrochemical, Quantum Chemical Studies and Synergistic Effect with Iodide ions. *Molecules*, 20:16004-16029

Wawrzkiewicz, M. & Hubicki, Z. (2009) Kinetics of adsorption of sulphonated azo dyes on strong basic anion exchangers. *Environ Technol*. 30:1059-71.

Wu, J. S., Liu, C. H., Chu, K. H. & Suen, S. Y. (2008). Removal of cationic dye methyl violet 2B from water by cation exchange membranes. *Journal of Membrane Science*, 309: 239-245.

Xue, H. Y., Han, F. & He, B. Y. (2012) Biosorption of food dye onto gelatin microspheres: Equilibrium isotherm and thermodyanamic analysis. *Advanced Materials Research*, 476-478: 2239-2242.

Yang, X. & Al-Duri, B. (2005) Kinetic modeling of liquid-phase adsorption of reactive dyes on activated carbon. *J. of Colloid Interface Science*, 287: 25-34.

Zhang, Y., Zhang, X., Lu, X., Yang, J. & Wu, K. (2010). Multi-wall carbon nanotube film-based electrochemical sensor for rapid detection of ponceau 4R and Allura red. *Food Chem*., 122: 909-913.

Zhang, Y., Zhou, H., Wang, Y., Wu, X. & Zhao, Y. (2015) Simultaneous determination of seven synthetic colorants in wine by dispersive micro-solid-phase extraction coupled with reversed-phase high-performance liquid chromatography. *J Chromatogr Sci*. 53:210-8.