TEKSTİL VE MÜHENDİS

(Journal of Textiles and Engineer)

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Comparison of Textile Dye Adsorption Properties of Low-Cost Biowaste Adsorbents

Düşük Maliyetli Biyolojik Atık Adsorbentlerin Tekstil Boyası Adsorpsiyonu Özelliklerinin Karşılaştırılması

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Online Erişime Açıldığı Tarih (Available online): 30 Eylül 2017 (30 September 2017)

Bu makaleye atıf yapmak için (To cite this article):

Fatma Oğuz ERDOĞAN (2017): Comparison of Textile Dye Adsorption Properties of Low-Cost Biowaste Adsorbents**,** Tekstil ve Mühendis, 24: 107, 181-187.

For online version of the article: https://doi.org/10.7216/1300759920172410706

TMMOB Tekstil Mühendisleri Odası
UCTEA Chamber of Textile Engineers
Tekstil ve Mühendis
Journal of Textiles and Engineer

Yıl (Year) : 2017/3
Cilt (Vol) : 24
Sayı (No) : 107

Araştırma Makalesi / Research Article

COMPARISON OF TEXTILE DYE ADSORPTION PROPERTIES OF LOW-COST BIOWASTE ADSORBENTS

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Gönderilme Tarihi / Received: 12.04.2017
Kabul Tarihi / Accepted: 04.08.2017

ABSTRACT: In this study, cherry and apricot stones have been investigated as an adsorbent for removing disperse yellow 211 textile dye from aqueous solution. Cherry and apricot stones have been characterized by SEM (scanning electron microscopy) and N_2 adsorption isotherms. In the study, the effects of adsorbent dosage, contact time, temperature and pH value on the removal efficiency have been investigated. The Freundlich, Langmuir, Temkin and Dubinin-Radushkevich (DR) isotherms were tested to examine the adsorption behavior. The kinetic studies indicate that adsorption follows the pseudo first order model. Thermodynamic parameters, enthalpy, entropy and Gibbs free energy changes were established. The maximum adsorption capacity was found to be as high as 105.71 mg/g for cherry stones and 156.25 mg/g for apricot stones. These results showed that the adsorption capacity of apricot stones is about 1.5 times higher than that of cherry stones. The results support the potential use of cherry and apricot stones as efficient and low-cost adsorbents for the removal of textile dye from aqueous solution.

Keywords: Apricot Stones, Cherry Stones, Textile Dye, Isotherm, Kinetic, Thermodynamic.

DÜŞÜK MALİYETLİ BİYOLOJİK ATIK ADSORBENTLERİN TEKSTİL BOYASI ADSORPSİYONU ÖZELLİKLERİNİN KARŞILAŞTIRILMASI

ÖZET: Bu çalışmada, kiraz ve kayısı çekirdekleri, sulu çözeltiden dispers sarı 211 tekstil boyasının uzaklaştırılması için bir adsorbent olarak incelenmiştir. Kiraz ve kayısı çekirdekleri SEM (taramalı elektron mikroskopisi) ve N_2 adsorpsiyon izotermleriyle karakterize edilmiştir. Çalışmada; adsorbent miktarı, temas süresi, sıcaklık ve pH'ın uzaklaştırma verimine etkisi incelenmiştir. Adsorpsiyon davranışının incelenmesinde Freundlich, Langmuir, Temkin ve Dubinin-Radushkevich (DR) izotermleri test edilmiştir. Kinetik çalışmalar, adsorpsiyonun yalancı birinci derece kinetik modeli takip ettiğini göstermektedir. Termodinamik parametreler olan entalpi, entropi ve Gibbs serbest enerji değişimleri belirlenmiştir. Maksimum adsorpsiyon kapasitesi kiraz çekirdekleri için 105.71 mg/g, kayısı çekirdekleri için 156.25 mg/g olarak bulunmuştur. Bu sonuçlar kayısı çekirdeklerinin adsorpsiyon kapasitesinin kiraz çekirdeklerinin adsorpsiyon kapasitesinden yaklaşık 1.5 kat fazla olduğunu göstermiştir. Sonuçlar, kiraz ve kayısı çekirdeklerinin, sulu çözeltiden dispers sarı 211 tekstil boyasının uzaklaştırılmasında etkili ve düşük maliyetli adsorbentler olarak kullanılma potansiyelini desteklemektedir.

Anahtar Kelimeler: Kayısı Çekirdekleri, Kiraz Çekirdekleri, Tekstil Boyası, İzoterm, Kinetik, Termodinamik.

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DOI: *10.7216/1300759920172410706, www.tekstilvemuhendis.org.tr*

1. INTRODUCTION

Dyes are environmental concerns because of their nonbiodegradability and toxicity. Many physico-chemical methods have been employed to remove dyes from aqueous solution, such as membrane filtration, chemical oxidation, ozonation, ion-exchange and adsorption. Among these methods, adsorption is considered to be the most efficient method [1-3]. Activated carbons are the most widely used adsorbents, but their high cost limits their commercial application. There are some approaches to find low cost and efficent, locally available adsorbents for the removal of dyes. Many studies have been reported use of agricultural or industrial wastes as dyes adsorbents which include: rice husk, papaya seeds, papaya leaves, hazelnut shell, wood sawdust, pineapple stem, banana leaves, bagasse pith, maize cob, fruit peels and coffee waste. There are some advantages of using agricultural and industrial waste materials for the removal of textile dyes because of saving disposal costs and preventing environmental problems. [4-7].
The world's largest producer of cherries (520000 tonnes) and apricots (811609 tonnes) is Turkey [8]. There is no effective area using these fruit waste. To the best of our knowledge, this is the first study investigates the adsorbent capacities of apricot and cherry stones for the removal of disperse yellow 211. The molecular structure of DY 211 was given in Figure 1.

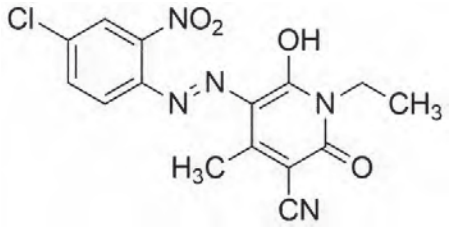

Figure 1. Chemical structure of Disperse Yellow 211 (DY 211).

The main objects of this paper are: (i) to study the potential use of apricot and cherry stones as an adsorbent for the removal of a textile dye, disperse yellow 211, (ii) to determine the effects of various parameters (i.e. pH, adsorbent dossage, temperature and contact time) on disperse yellow 211 adsorption, (iii) to determine the conformity of adsorption data to Langmuir, Freundlich, Temkin and DR models to find out the optimal isotherm equation, and (iv) to determine kinetic and thermodynamic parameters and explain the adsorption behavior. Moreover, a comparison between apricot stones and cherry stones for adsorption of dye was done.

2. MATERIAL AND METHODS

Apricot stones (AS) and cherry stones (CS) used in this study were obtained locally. The fruit stones (apricot or cherry) were dried, crushed, and sized to between 1 and 3 mm, then treated with 15% H_2SO_4 for 12 h and washed with hot distilled water. The surface morphology was examined using SEM (Jeol/Jsm-6335 F-Inca/Eds). The BET surface areas of apricot and cherry stones were determined using nitrogen at 77.4 K (Quantachrome Autosorb iQ).

2.1. Adsorption Equilibrium Studies

Adsorption of DY 211 onto the low-cost adsorbents has been studied by batch experiments. In experimental studies; different amounts of adsorbents (in the range of 0.02-0.035 g/L) have been used. The initial concentrations and the solution volumes were setted to 140 mg/L and 20 mL, respectively. Flasks containing adsorbents and DY 211 solutions, were shaken in a mechanical shaker (GFL 1086) at 100 rpm and different temperature (30, 40 and 50°C). After adsorption process, samples were filtered and the concentrations of disperse yellow 211 in the solutions were determined using UV spectrophotometer (LaboMed Inc.) at 491 nm. The adsorption efficiency, *E*, was calculated using Eq. (1):

$$E = \frac{C_0 - C_e}{C_0} \cdot 100 \quad (1)$$

where C_0 is the initial liquid phase concentration of DY 211 and C_e is the equilibrium liquid phase concentration of DY 211, both C_0 and C_e is in mg/L. The disperse yellow 211 uptake at equilibrium, q_e (mg/g), was calculated using Eq. (2):

$$q_e = \frac{(C_0 - C_e)V}{W} \quad (2)$$

where *V* (L) is the solution volume, and *W* (g) is the mass of adsorbent used [9].

The effect of pH on the adsorption of DY 211 was examined by carrying out the adsorption process at different pHs. Adsorbent (cherry or apricot stones) dosage of 0.02 g/L and adsorption temperature of 50 °C. The pH was adjusted by adding a few drops of diluted 0.1 N NaOH or 0.1 N HCl before each experiment and measured using a pH meter (Crison pH 25). The equilibrium data were simulated using the Freundlich, Langmuir Temkin and DR isotherm models.

2.3. Adsorption Kinetics

For the kinetic studies, experiments have been carried out by adding 0.02 g/L adsorbent to 20 mL of 140 mg/L DY 211 solutions at 303, 313 and 323 K at optimum pH. The DY 211 uptake, q_t (mg/g), was calculated by the following equation:

$$q_t = \frac{(C_0 - C_t)V}{W} \quad (3)$$

where C_t (mg/L) is the liquid-phase concentration of DY 211 at the time t (min).

In order to examine the adsorption behavior of DY 211, pseudo first order, pseudo-second order and intra-particle diffusion kinetics equations which are described in Eqs. (4), (5) and (6), respectively, were applied for modeling experimental data [6, 10-11]

$$log(q_e - q_t) = logq_e - \frac{k_1 t}{2.303} \quad (4)$$

$$\frac{t}{q_1} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (5)$$

$$q_t = k_i t^{1/2} + C \quad (6)$$

where k_1 (1/min), k_2 (g/mg min) and k_i (mg/g min1/2) are the adsorption rate constants of the pseudo first order, pseudo second order and intra-particle diffusion, respectively. C (mg/g) is a constant.

3. RESULTS

3.1. Characterization of Adsorbents

The surface physical properties of adsorbents were characterized with an automated gas sorption apparatus using N_2 as adsorbate at 77.4 K. Nitrogen adsorption is a standart technique widely used for the determination of porosity of adsorbent. Table 1 shows the BET and Langmuir surface areas, total pore volume and avarage pore size for the adsorbents. The BET and Langmuir surface areas of the cherry stones were 17.67 m^2/g and 23.85 m^2/g, respectively.

Table 1. Textural parameters of the adsorbents.

Adsorbent	Cherry Stones	Apricot Stones
BET Surface Area (m^2/g)	17.67	7.42
Langmuir Surface Area (m^2/g)	23.85	11.25
Total Pore Volume (mL/g)	6.78E-3	3.35E-3

The BET and Langmuir surface areas of the cherry stones are larger than the surface areas of apricot stones. The total pore volume of cherry stones and apricot stones were 6.78 E-3 and 3.35 E-3 mL/g, respectively. SEM was used to observe the surface physical morphology of the cherry stones and apricot stones. SEM image of the apricot stones is shown in Fig. 2. SEM image of raw cherry stones was reported in our previous study (Fig. 5) [7]. The surface of cherry stones and apricot stones has very small pores.

3.2. Batch Adsorption Studies

The curve adsorption uptake of DY 211, q_t as a function of time, t at the amounts of adsorbent weights in the range of 0.02-0.035 g/L is displayed in Fig.3. The adsorption capacity increased with prolonging the contact time for two adsorbents.

It is clear from Fig.3 that the adsorption process increased drammatically at the initial stage, indicating the availability of readily accessible sites. The adsorption process is gradually slowing down as it approaches the equilibrium. The time required to reach the equilibrium state is called as equilibrium time [6]. Fig. 3 showed that the adsorption capacities at equilibrium (q_e) decreased with an increase in the amount of adsorbent from 0.02 to 0.035 g/L for both adsorbents. This is explained as a consequence of partial aggregation, which occurs at high adsorbent amount resulting in decreased active sites. A similar phenomenon was reported by Huang et al. [9], Rubin et al. [12], and Nweke and Okpokwasili [13]. In a batch of adsorption studies, the adsorption efficiency (E) increased from 68.02% to 80% when the adsorption dosage of apricot stones increased from 0.02 to 0.035 g/L at 30 °C. Similar results have also been observed in other temperatures and adsorbent (cherry stones). This finding agrees with the recent work by Huang et al. [9] and Kosa et al. [14].

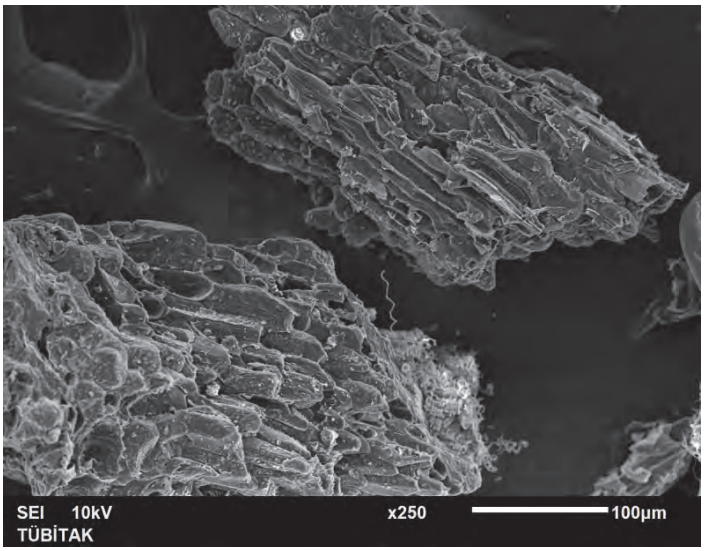

Figure 2. SEM image of the apricot stone (AS).

The effect of pH was investigated by varying the pH of dye solutions from 3 to 11. As can be seen from Fig 4, increasing in the solution pH from 3 to 11 caused an increasing in q_e from 40.15 to 80.72 mg/g for apricot stones-based adsorbent (AS). At strong acidic pHs, itr was obserbed that the amount of adsorption decreased, probably due to the presence of H_3O^+ ions competing with DY 211 cations for the adsorption sites. At basic pHs, the apricot stones-based adsorbent may get negatively charged and the formation of electric double layer changes its polarity and consequently the DY 211 dye uptake increases. A similar phenomenon was reported by Krishni et al. [6]. Figure 4 shows the dependency of DY 211 adsorption on cherry stones-based adsorbent (CS) on pH. At acidic and basic pHs, the adsorption capacity of DY 211 getting higher than at neutral pHs. This situation can be explained by the formation of positive and negative charges in acidic and basic solutions, respectively. This result corroborates the reports of our previous study [2].

The temperature dependence of DY 211 sorption onto CS and AS was studied at the optimum adsorbent dosage of 0.02 g/L and pH 11. As can be seen in Figure 5 the DY 211 adsorption increased with the increasing temperature for both adsorbents. This situation can be explained by the increasing reaction rate at higher temperatures. Another possible explanation is that high temperature extends the pore volume and surface area and provides more chances for DY 211 to pass the external boundary layer and penetrate more easily. This corroborates the reports of

our previous study [2]. Similar behavior was reported in the literature [9,15-16].

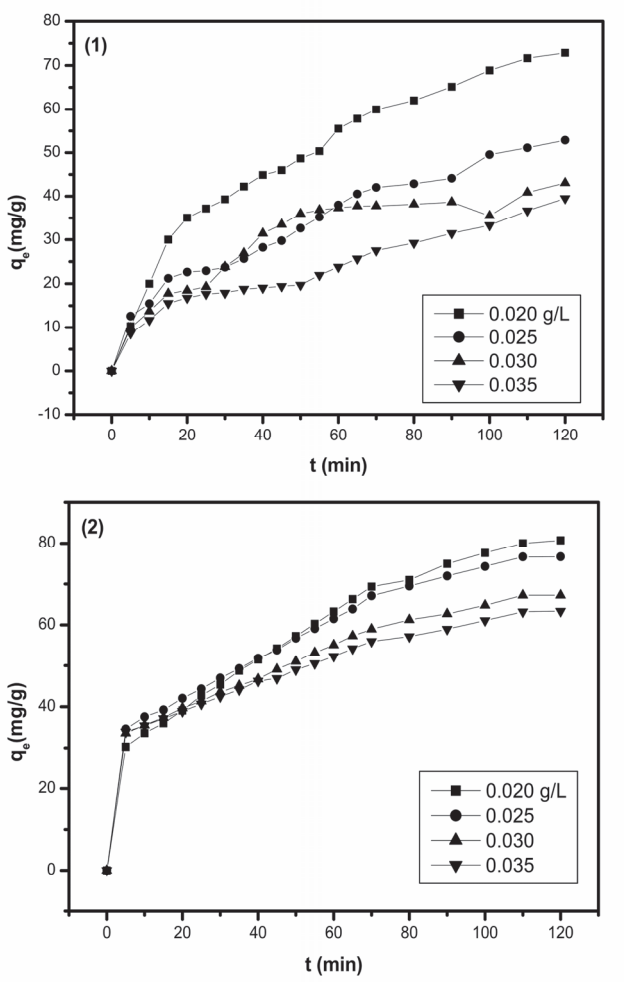

Figure 3. Effects of adsorbent dosage and contact time on the uptake of textile dye onto the CS (1) and AS (2) (conditions: C_0=150 mg/L; temperature=50 °C).

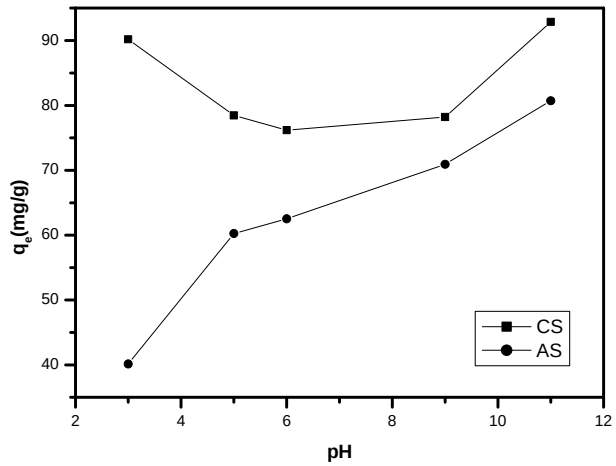

Figure 4. Effect of pH on the adsorption of textile dye onto CS and AS (conditions: W=0.020 g/L; C_0=150 mg/L).

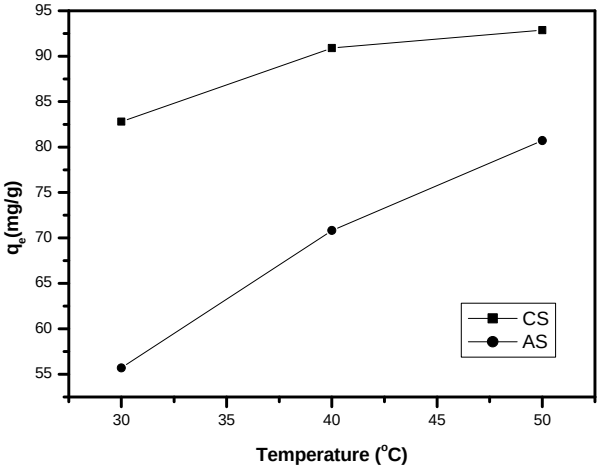

Figure 5. Effect of solution temperature on the adsorption textile dye onto CS and AS (conditions: W=0.02 g/L; C_0=150 mg/L).

3.3. Adsorption Isotherms

The equilibrium adsorption isotherms provides a practical approach for design and optimization of adsorption processes. Adsorption isotherms describe the interaction between adsorbates and adsorbents. [5]. The equilibrium data for textile dye adsorption onto cherry or apricot stones were explored using the isotherm model of Langmuir, Freundlich, DR and Temkin. The parameters obtained from the four isotherm models were represented in Table 2. T he correlation coefficients of cherry stones was found to be higher than apricot stones. Langmuir adsorption capacities, Q_0, were found as 105.71 and 156.25 mg/g for cherry and apricot stones, respectively. The R_L values were found to be 0.117 and 0.472 for cherry and apricot stones, respectively, indicating that the adsorption is favorable ($0<R_L<1$). Estimated free energies from DR model were found to be 7.245 and 6.399 kj/mol for the cherry and apricot stones, respectively. Free energies below 8 kj/mol corresponds to the adsorption process of physical nature [17].

Table 2. Freundlich, Langmuir, Temkin and DR isotherm constants for the adsorption of DY 211 onto cherry and apricot stones-derived adsorbent at 30 °C.

Adsorbent	**Isoterms and Parameters**		
	Freundlich		
	K_F (mg/g)(L/mg)$^{1/n}$	n	R^2
CS	22.12	3.14	0.944
AS	2.05	1.274	0.836
	Langmuir		
	Q_0(mg/g)	K_L (L/mg)	R^2
CS	105.71	0.0538	0.937
AS	156.25	0.0080	0.784
	Temkin		
	A (L/g)	B	R^2
CS	0.567	23.04	0.944
AS	0.061	40.26	0.811
	DR		
	qm(mg/g)	E(kJ/mol)	R^2
CS	105.5	7.245	0.893
AS	84.42	6.399	0.947

3.4. Kinetics of Adsorption

Adsorption kinetics provides information for the adsorption mechanisms. Textile dye, DY 211 was adsorbed on cherry and apricot stones as a function of time. Pseudo first and second order and also intraparticle diffusion model have been obtained at 30, 40 and 50 °C for various amounts of adsorbent (0.02-0.035 g/L). The regression coefficients (R^2) were evaluated for all models. Kinetic parameters are represented in Table 3 and 4.

As shown in Table 3 and 4, the hightest R^2 values were obtained for the pseudo first order kinetic model and the experimental q_e values matched well with the calculated data. In Table 3 and 4, it can be seen that k_1 values are decreasing with the increasing amount of adsorbent at 30, 40 and 50 °C. The average values for k_1 are 0.0316, 0.0251 and 0.0247 at 30, 40 and 50 °C, respectively for cherry stones. Despite the high R^2 values obtained from the pseudo first and second order kinetic models, the q_e values obtained from the pseudo first order model showed better agreement with the experimental results. Therefore, it can be said that the pseudo second order model is not suitable to explain the adsorption process accurately. Similar results have been reported for the sorption kinetics of various adsorbate onto various adsorbents in literature [12, 18-19]. Kinetic parameters for the removal of DY 211 by apricot stones are represented in Table 4. The results for the apricot stones showed good agreement with the three kinetic models, especially with the pseudo first order kinetic model. The average values for k_1 are 0.0273, 0.0246 and 0.025 at 30, 40 and 50 °C, respectively for apricot stones. This behaviour was also reported by Nesic et al. [11], Kondapalli and Mohanty [18], Agarry and Aremu [20], Varshini and Das [21], Akar et al. [22].

Table 3. Kinetic models parameters for the adsortion of DY 211 onto cherry stones (CS) at different temperature and adsorbent dosage.

			Pseudo first order		**Pseudo second order**			**Intraparticle diffusion**		
Temperature (°C)	m_{ads} (g/L)	$q_{e,exp}$ (mg/g)	k_1 $(min)^{-1}$	$q_{e,cal}$ (mg/g)	R^2	k_2 (g/mg.min)	$q_{e,cal}$ (mg/g)	R^2	k_p	R^2
30	0.02	90.87	0.0414	89.54	0.993	3.08E-4	109.05	0.973	8.322	0.963
	0.025	76.69	0.0318	80.75	0.969	3.34E-4	94.34	0.957	6.863	0.982
	0.030	68.57	0.0297	44.53	0.939	9.89E-4	75.87	0.997	5.866	0.857
	0.035	64.41	0.0237	67.04	0.935	3.48E-4	76.75	0.907	5.416	0.974
40	0.02	82.87	0.0296	84.60	0.913	3.74E-4	97.94	0.982	7.300	0.980
	0.025	79.89	0.0270	80.33	0.911	3.18E-4	95.51	0.944	6.852	0.982
	0.030	71.24	0.0255	56.85	0.955	5.20E-4	83.26	0.984	6.403	0.937
	0.035	66.70	0.0183	60.23	0.996	3.80E-3	75.08	0.894	5.268	0.959
50	0.02	92.87	0.0299	86.28	0.914	4.56E-4	104.9	0.985	7.715	0.961
	0.025	87.89	0.0258	81.67	0.912	3.80E-4	100.7	0.959	7.187	0.968
	0.030	77.90	0.0247	53.80	0. 973	6.67E-4	85.84	0.982	6.537	0.907
	0.035	72.41	0.0185	61.04	0.995	4.44E-4	78.93	0.919	5.505	0.949

Table 4. Kinetic model parameters for the adsortion of DY 211 onto apricot stones (AS) at different temperature and adsorbent dosage.

			Pseudo first order			**Pseudo second order**			**Intraparticle diffusion**	
Temperature (°C)	m_{ads} (g/L)	$q_{e,exp}$ (mg/g)	k_1 $(min)^{-1}$	$q_{e,cal}$ (mg/g)	R^2	k_2 (g/mg.min)	$q_{e,cal}$ (mg/g)	R^2	k_p	R^2
30	0.02	55.72	0.0436	60.252	0.999	7.58E-4	78.80	0.989	5.644	0.919
	0.025	48.68	0.0235	40.361	0.915	7.63E-4	53.02	0.976	3.697	0.949
	0.030	43.91	0.0227	40.454	0.958	4.71E-4	51.02	0.951	3.549	0.968
	0.035	40.50	0.0194	35.68	0.989	5.54E-4	48.50	0.983	3.550	0.989
40	0.02	70.72	0.0283	74.40	0.923	3.08E-4	87.26	0.946	6.299	0.984
	0.025	68.69	0.0260	74.07	0. 978	1.16E-4	110.99	0.937	6.868	0.985
	0.030	60.58	0.0252	66.23	0.984	1.85E-4	89.21	0.955	5.993	0.984
	0.035	57.64	0.0188	50.70	0.990	3.73E-4	69.54	0.972	5.085	0.989
50	0.02	80.72	0.0287	102.98	0.980	2.39E-5	228.3	0.720	8.783	0.965
	0.025	76.68	0.0286	103.0	0.980	5.32E-5	154.56	0.814	7.970	0.961
	0.030	67.24	0.0240	81.401	0. 944	9.51E-5	113.9	0.833	6.642	0.968
	0.035	63.35	0.0187	62.96	0.984	2.08E-4	84.25	0.929	5.705	0.996

3.5. Adsorption Thermodynamics

In describing the thermodynamic behavior of the adsorption of DY 211 onto cherry or apricot stones, thermodynamic parameters (i.e ΔG°, ΔH° and ΔS°) were calculated with Eq. 7 and 8.

$$\Delta G = -RTlnK_c \quad (7)$$

$$\ln K_c = -\frac{\Delta H^\circ}{RT} + \frac{\Delta S^\circ}{R} \quad (8)$$

where, T is the temperature in Kelvin, R is the universal gas constant (8314 J/kmol K) and K_C is the equilibrium constant (C_{ads}/C_e) [3]. ΔH° and ΔS° were determined from the $\ln K_C$ against 1/T plot. Negative ΔG° values showed that the adsorption of DY 211 is spontaneous and feasible. Increasing in the magnitude of ΔG° with the increasing temperature, means that the adsorption becomes more feasible at higher temperatures. [9, 18, 23] In general, ΔG° values between 0 and -20 kJ/mol corresponds to physical adsorption, and the chemical adsorption occurs when the ΔG° value is in the range of -80 to -400 kJ/mol [9]. The positive value of ΔH° indicates an endothermic process which is supported by the increased adsorption of DY 211 with increasing temperature. The magnitude of ΔH° is very useful in describing the type of adsorption. If the magnitude of ΔH° is in the range of 2.1-20.9 kJ/mol, it denotes a physical adsorption process, while chemisorptions fall within the range of 80-200 kJ/mol [18, 19].

From Table 5 and 6, the value of ΔH° obtained for both adsorbents showed a physical adsorption process. This was also supported by DR isotherm results (E<8 kJ/mol) for both adsorbents. This explains why the q_e values obtained from the pseudo second order kinetic model (chemisorptions model) did not show a good agrement with the experimental data. Furthermore, for both adsorbents, positive ΔS° values correspond to the increased randomness at the solid-solution interface during the adsorption process.

Table 5. Thermodynamic parameters for the adsortion of DY 211 onto cherry stones-derived adsorbent

T (°C)	ΔG° (kJ/mol)	ΔH°(kJ/mol)	ΔS° (J/mol)	R^2
30	-0.90	12.412	44.143	0.907
40	-1.55			
50	-1.77			

Table 6. Thermodynamic parameters for the adsortion of DY 211 onto apricot stones-derived adsorbent

T (°C)	ΔG° (kJ/mol)	ΔH°(kJ/mol)	ΔS° (J/mol)	R^2
30	-0.018	11.626	38.50	0.999
40	-0.389			
50	-0.787			

4. DISCUSSION AND CONCLUSION

Cherry and apricot stones were used as low-cost adsorbents for the removal of DY 211 from aqueous solutions at various temperatures. In adsorption studies, the efficiency of DY 211 adsorption by cherry or apricot stones increased with adsorbent dosage, but the equilibrium adsorption capacity decreased significantly. The results showed that the adsorption capacity of apricot stones is about 1.5 times higher than that of cherry stones. The adsorption capacity of both adsorbents for DY 211 increased with the increasing temperature. In the assessment of experimental data, various kinetic models were tested to understand the adsorption mechanism. The results showed that the pseudo first order kinetic model exhibits good agreement with the dynamic behavior of the DY 211 adsorption onto the both adsorbents. For the mathematical description of the adsorption, Freundlich, Langmuir, Temkin and DR isotherm models have been used. The results showed that the adsorption equilibrium data fitted well to the Freundlich and Temkin models for cherry stones and DR and Freundlich models for apricot stones. Thermodynamic parameters (i.e. ΔG°, ΔH° and ΔS°) were also determined. In all cases, obtained ΔG° values showed that the adsorption of DY 211 is spontaneous and feasible, positive ΔH° values indicates that the adsorption process is endothermic and the positive ΔS° values indicates the increasing randomness of the adsorption process for both adsorbents. This study has revealed that cherry and apricot stones can be used as an efficient and economically viable adsorbent for removal of DY 211 from aqueous solutions.

ACKNOWLEDGMENT

The author acknowledge the financial support provided by Kocaeli University Scientific Research Projects Unit. (Project No: 2014/113 HDP, 2016/019 HD). The author thanks to HÜBTUAM (Hitit University) for the surface area measurements.

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