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Research Article

Investigation of the Use of Activated Peach Kernel in Cr(VI) Adsorption from Aqueous Solution

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Abstract

Heavy metals're threats to aquatic ecosystems. Heavy metal-containing wastewater causes environmental pollution as a result of reaching the aquatic ecosystem after being used by industries. In this study, the use of activated peach kernel (APK) as a bioadsorbent and its use in Cr(VI) adsorption from aqueous solution were investigated. Thus, it's aimed to increase the economic value of these materials, which have low economic value, by investigating the possibilities of using them as bioadsorbent. Other hand, it's aimed to contribute to the protection of both the aquatic environment and human health, and thus to the prevention of environmental pollution, by removing heavy metals. In the study, synthetically wastewater containing Cr(VI) was used. Atomic Adsorption Spectrophotometer (AAS), Fourier Transform Infrared Spectroscopy (FT-IR), Brunauer-Emmett-Teller Isotherm Model (BET) and Scanning Electron Microscope (SEM) were used to determine the Cr(VI) concentration and characterization of the adsorbent. From the adsorption processes performed after the activation processes, the yield with APK was 93%. In the study, maximum efficiency was reached at pH 2, 0.5 g adsorbent amount and 5 hours retention time under optimum conditions. It was observed that 0.738 $mg.g^{-1}$ removal capacity per unit adsorbent was obtained with 0.5 g APK under optimum conditions. It was found to be suitable for the Langmuir isotherm model, which is one of the adsorption isotherm models. In the kinetic evaluation of the reaction, it was concluded that it was suitable for pseudo-second-order kinetics. All results demonstrated the usability of APK in Cr(VI) removal, especially due to activation process.

Keywords: Cr(VI), Peach kernel, Adsorption, Bioadsorbents, Langmuir

Aktifleştirilmiş Şeftali Çekirdeğinin Sulu Çözeltiden Cr(VI) Adsorpsiyonunda Kullanımının Araştırılması

Öz

Ağır metaller, çevresel açıdan su ekosistemlerine yönelik tehditlerden biridir. Ağır metal içeren atıksular endüstriler tarafından kullanıldıktan sonra sucul ekosisteme ulaşması sonucunda çevre kirliliğine neden olmaktadır. Bu çalışmada aktifleştirilmiş şeftali çekirdeğinin (APK) biyoadsorbent olarak kullanımı ve sulu çözeltiden Cr(VI) adsorpsiyonunda kullanılabilirliği araştırılmıştır. Böylece ekonomik değeri düşük olan bu malzemelerin biyoadsorbent olarak kullanım olanakları araştırılarak ekonomik değerlerinin arttırılması amaçlanmıştır. Öte yandan, sucul ekosistemlerde kirletici tehditler olan ağır metallerin bu malzemelerle giderilerek hem sucul ortamın hem de insan sağlığının korunmasına ve dolayısıyla çevre kirliliğinin önlenmesine katkı sağlanması hedeflenmektedir. Çalışmada Cr(VI) içeren sentetik atıksu kullanılmıştır. Adsorbentin Cr(VI) konsantrasyonunu ve karakterizasyonunu belirlemek için Atomik Adsorpsiyon Spektrofotometresi (AAS), Fourier Dönüşümü Kızılötesi Spektroskopisi (FT-IR), Brunauer-Emmett-Teller İzoterm Modeli (BET) ve Taramalı Elektron Mikroskobu (SEM) kullanılmıştır. Aktivasyon işlemlerinden sonra gerçekleştirilen adsorpsiyon işlemlerinden APK ile verimi %93 olmuştur. Çalışmada optimum koşullar altında pH 2, 0.5 g adsorbent miktarı ve 5 saat alıkonma süresinde maksimum verime ulaşılmıştır. Optimum koşullar altında 0.5 g APK ile birim adsorbent başına 0,738 $mg.g^{-1}$ kaldırma kapasitesi elde edildiği gözlemlenmiştir. Adsorpsiyon izoterm modellerinden Langmuir izoterm modeline uygun olduğu görülmüştür. Reaksiyonun kinetik değerlendirmesinde yalancı ikinci dereceden hız kinetiğine uygunduğu sonucuna ulaşılmıştır. Tüm sonuçlar, özellikle aktivasyon süreci nedeniyle, Cr(VI) gideriminde APK'nin kullanılabilirliğini ortaya koymuştur.

Anahtar Kelimeler: Cr(VI), Şeftali çekirdeği, Adsorpsiyon, Bioadsorbent, Langmuir

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1. Introduction

A wide variety of chemical pollutants released as a result of industrial and agricultural activities have become an important issue affecting environmental safety. People are exposed to several pollutants accumulated in the environment because of such activities. One of the pollutions affecting environmental systems is water pollution. Hazardous and other wastes from industries are discharged directly or indirectly to water resources (Shouli, ve diğerleri, 2010) (Gupta, Rastogi, & Nayak, 2010) (Gupta & Rastogi, 2009a) (Gupta, Goyal, & Sharma, 2009b). Heavy metals are naturally occurring elements with atomic weight and density at least 5 times higher than the water atom (Tchounwou, Yedjou, Patlolla, & Sutton, 2012). It causes the distribution of heavy metals in large areas in the environment, originating from many industrial, domestic, agricultural, medical, and technology applications.

Considering the effects of heavy metal pollution on human health and the environment, it is seen that the toxicity effect increases according to the exposure time. Considering the factors causing the toxicity effect, it is seen that various factors such as dose, exposure route, and chemical species as well as age, gender, genetics, and nutritional status of the exposed persons are effective. Due to their high toxicity, arsenic, cadmium, chromium, lead, and mercury are among the priority heavy metals that are important for public health. These metallic elements appear to cause multiple organ damage even at lower exposure levels (Bradl, 2002). They are also classified as human carcinogens (known or probable) according to the US Environmental Protection Agency (EPA) and the International Agency for Research on Cancer (IARC) (Tchounwou, Yedjou, Patlolla, & Sutton, 2012).

Chromium is released into the environment not only from natural sources but also as a result of human activities. The main source of chromium pollution is mining; however, it also appears in various industries such as cement ceramics, glass industries, paint industry, zinc coating industries, production of steel and other metal alloys, photographic material, and rust-removing paints (Rana, N., & Rajagopal, 2004) (Rutland, 1991). Although Cr (III) is non-toxic or less toxic than Cr(VI), prolonged exposure to trivalent chromium is known to cause allergic skin reactions (Gupta & Rastogi, 2009a). Since Cr(VI) is toxic and carcinogenic, it must be treated before being discharged to wastewater (JParga, ve diğerleri, 2005). Conventional methods such as membrane filtration (Fu & QiWang, 2011), chemical precipitation (Ku & Jung, 2001), ion exchange (Kang, Le, Moon, & Kim, 2004), and adsorption (Fu & QiWang, 2011) are using to remove chromium from wastewater.

Although various treatment methods are using for the removal of Cr(VI), heavy metal removal is possible by using the adsorption method, which is another advanced treatment method. The adsorption method is acceptable as an effective and economical method for heavy metal wastewater treatment. The adsorption method is a simple method to use and an effective method for heavy metal removal. Adsorption processes can be reversible in some cases, in which case the adsorbents can be regenerated by appropriate desorption processes (Fu & QiWang, 2011).

The most important disadvantage of using the adsorption method for chrome removal from wastewater is the high production cost of adsorbents. Instead of using adsorbents with high production costs, it is possible to reduce costs by using naturally obtained and usable natural adsorbents (Gupta, Ali, Saleh, Siddiqui, & Agarwal, 2013).

2. Material and Method

2.1. Material

Peach kernels were used in the study. The peach kernel was taken from the juice processing factory. The core pulp in the peach kernel was separated and the hard part outside was used in the experiments.

2.2. Methods

2.2.1. *Preparation of the Adsorbent*

The adsorbents used during the experiments were first washed with distilled water and the dust and unknown substances in it were cleaned and then dried in an oven at 105 °C for 24 hours. The dried adsorbent was grinded to the desired size and then sieved with a 35-mesh sieve gap.

1 M Hydrochloric Acid (HCl) solution was prepared for the activation of adsorbents. 10 g adsorbent was added to the prepared HCl solution, and the solution was mixed at 150 rpm for 24 hours. The adsorbents taken from the shaker were filtered with filter papers and dried in an oven at 105 °C for 24 hours. After the processes, adsorption experiments were performed with activated peach kernel (APK).

2.2.2. *Preparation of Stock Solutions*

Chromium, a heavy metal, was prepared by dissolving Sigma-Aldrich brand $K_2Cr_2O_7$ of distilled water for a stock solution. The stock solution was stored at room conditions throughout the experiments and the desired concentrations were prepared using this stock solution.

2.2.3. *Characterization of Adsorbent*

In adsorption experiments, ISOLAB precision balance was used to adjust the weighing amounts; Stuart biolab orbital shaker was used to mix the solutions at 150 rpm. ISOLAB pH meter was used to determine the acid-base degree, Heraeus Brand T-12 Model oven was used to dry the samples, and Perkin Elmer A Analyst 200 model Atomic Adsorption Spectrophotometer was used to measure the amount of chromium in the samples.

SEM and BET analyzes to examine the surface morphology of the adsorbents and to look at the adsorbent capacity before and after the adsorption experiments; FTIR analyzes were applied to determine the infrared spectra generated during the experiment (Saleh, Yalvaç, Arslan, & Gün, 2019).

2.2.4. *Adsorption Study*

pH optimization, initial Cr(VI) concentration, adsorbent dosage, and time optimizations were made for Cr(VI) removal from an aqueous solution. During the experiments, a volume of 100 ml was used and mixed at a constant stirring speed of 150 rpm in 250 ml beakers in case of overflow. In the experiments, 0.1 M HCl and 0.1 M NaOH were used for pH adjustments. As a result of the measurements, efficiency, and adsorbent capacity (qe) calculations are shown in Equations 1 and 2 (Shakoor & Nasar, 2018).

$$Efficiency\ \% = \frac{(C_i - C_f)}{C_i} \times 100 \qquad \text{Equation 1}$$

$$q_e = \frac{(C_i - C_f) \times V}{m} \qquad \text{Equation 2}$$

Where, C_i is the initial concentration ($mg.L^{-1}$) of the Cr(VI); C_f is the Cr(VI) final concentration ($mg.L^{-1}$); V solution volume (L); m adsorbent mass (g).

2.2.5. *Adsorption Isotherm*

Adsorption isotherms are the set of parameters applied to understand the adsorption mechanisms (Ho & McKay, 1999). Commonly used isotherms are Freundlich and Langmuir equations (Sawyer & P.L., 1978). According to the Langmuir isotherm (Equation 3), the energy level on the adsorbent surface is the same everywhere. The film formed by holding the atoms or molecules by active centers on the adsorbent surface is considered mono film (Filiz, 2007). The Freundlich isotherm (Equation 4) is a type of reversible adsorption that occurs physically on a heterogeneous surface. It is used in multi-layer systems where the adsorption heat and affinity are not evenly distributed on the heterogeneous surface (Benefield, Judkins, & B.L., 1982).

$$\frac{C_A}{q_A} = \frac{1}{b_A q_m} + \frac{C_A}{q_m} \qquad \text{Equation 3}$$

$$\mathrm{Log}(q_A) = \log(K_A) + \left(\frac{1}{n}\right)\log(C_A) \qquad \text{Equation 4}$$

Where C_A is the concentration of adsorbate A at the equilibrium ($mg.L^{-1}$). b_A is the Langmuir constant for the adsorbate A ($L.mg^{-1}$). q_M maximum adsorbent capacity at saturation ($mg.g^{-1}$). K_A is the Freundlich adsorption capacity parameter ($mg.g^{-1}$) ($L.mg^{-1}$). 1/n is the intensity parameter.

2.2.6. *Adsorption Kinetics*

Adsorption kinetics is used to determine the time required for adsorption equilibrium to occur. Many kinetic models have been developed to detect the chemical reaction and mass transfer of adsorption. The pseudo-first-order kinetic model predicted by Lagergren (Pseudo-first-order kinetic model) (Equation 5) and the pseudo-second-order kinetic model predicted by Ho (Pseudo second-order kinetic model) (Equation 6) is the most common kinetic models (Metcalf & H.P., 2003) (Tosun, 2009) (Kocabaş, 2015).

$$Log(q_e - q_t) = Log q_e - \frac{K_1}{2.303} t \qquad \text{Equation 5}$$

$$\frac{t}{q_t} = \frac{1}{K_2 q_e^2} + \frac{1}{q_e} t \qquad \text{Equation 6}$$

In the equations, q_e is the amount of adsorbed material in equilibrium; qt, is the amount of adsorbed material at the end of time t; K_1 represents the first-order rate constant (min^{-1}) of adsorption and K_2 shows the false second-order rate constant ($g.mg^{-1}.h^{-1}$).

3. Results and discussion

3.1. Adsorbent characterization

In this study, characterization was carried out to understand the properties of the adsorbent. According to the SEM results, it can be considered that the adsorbent surface is heterogeneous. SEM images of the APK structure before and after the adsorption process are shown in the photos below. Figure 1.a raw APK, b shows APK after adsorption, while APK is shown.

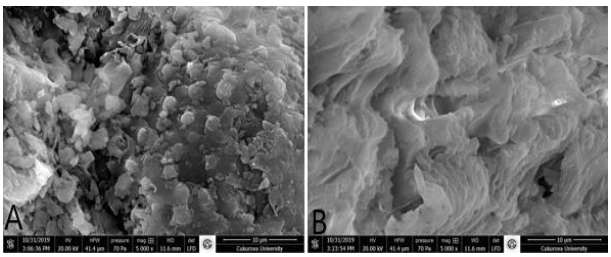

Figure 1. a) SEM for APK before adsorption b) SEM for APK after adsorption

Şekil 1. a) APK adsorpsiyon öncesi SEM görüntüsü b) APK adsorpsiyon sonrası SEM görüntüsü

The surface area of the adsorbent was determined by BET analysis. Before the adsorption process, the BET surface area and Langmuir surface area for the adsorbent were 2.02 $m^2.g^{-1}$ and 3.29 $m^2.g^{-1}$.This value is approximately equal to the surface area of the Xanthium Italicum plant (Saleh, Yalvaç, & Arslan, 2019). The BET surface area and Langmuir surface area decreased to 0.65 and 0.83 $m^2.g^{-1}$, respectively, at the end of the adsorption.

Fourier transform infrared spectra for adsorbent APK were determined using FT-IR analysis. The adsorbance bands observed at 3328 and 2891 are assigned to the -OH and -CH_2 stretch, respectively. The peak associated with the C=N stretching functional group was noticed to be in a band of 1644 cm-1. The peak at 2111 cm^{-1} on CC stretching. Many hills have been changed. The peak at 1318 indicates O-H bond. These results confirm the effect of the functional group. Since the adsorbent structure changes, it can be considered as chemical adsorption. FT-IR analysis a) before adsorption b) after adsorption. It is given in Figure 2.

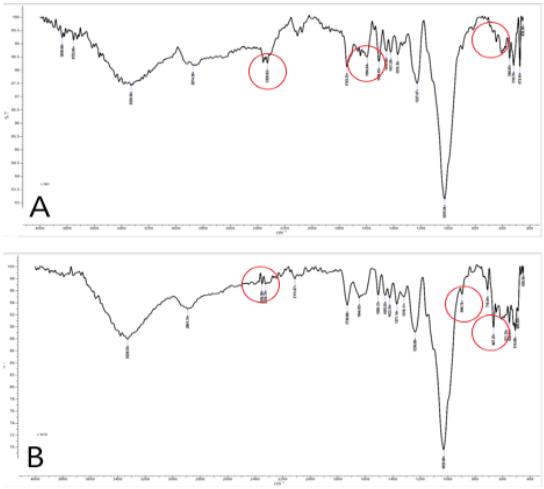

Figure 2. FTIR analysis a) APK before adsorption b) APK after adsorption

Şekil 2. FTIR analizi a) APK adsorpsiyon öncesi b) APK adsorpsiyon sonrası

3.2. Adsorption Studies

3.2.1. *Effect of pH*

The pH effect was investigated by adding APK waste to the wastewater containing Cr(VI) that we prepared in the study. The

pH effect, which affects both the adsorption surface and the adsorbent type, was investigated. To investigate the effect of pH on adsorption, other variables were kept constant and samples with pH values in the range of 2, 4, 6, 8, 10, respectively, were taken and adsorption studies were carried out using APK. The values obtained at the end of the adsorption studies lasting 24 hours by adding 1 g adsorbent to the sample containing 5 ppm Cr(VI) concentration are given in Figure 4.3. It was observed that the adsorption capacity of APK in wastewater containing Cr(VI) reached the highest removal efficiency at pH 2. It was observed that the lowest adsorption value was at pH 10. The effect of pH values on wastewater containing Cr(VI) is given in Figure 3.

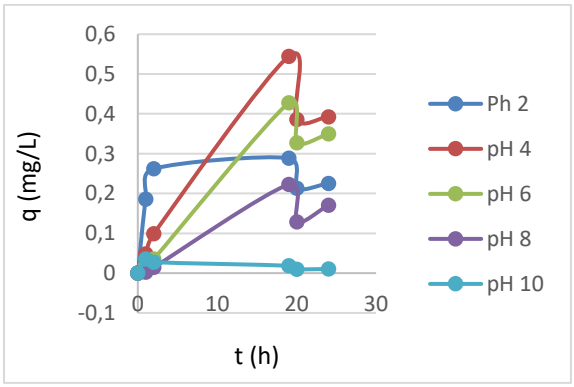

Figure 3. pH effect of APK on wastewater containing Cr(VI)

Şekil 3. pH'ın Cr(VI) içeren atıksu üzerine etkisi

3.2.2. *Effect of Adsorbent Amount*

To investigate the adsorbent effect on adsorption by adding synthetic wastewater containing Cr(VI) on APK, 0.3, 0.5, 1 g APK was used, and the results are given in Figure 4. It was observed that a removal capacity of 0.738 $mg.g^{-1}$ was obtained when 0.5 g of APK was added to the wastewater containing Cr(VI) given in Figure 4. It was observed that a removal capacity of 0.2757 $mg.g^{-1}$ was obtained when 1 g of APK was added to the wastewater containing Cr(VI). Figure 4. The effect of the amount of adsorbent on the Cr(VI)-containing wastewater is observed. The effect of the amount of adsorbent of APK on the wastewater containing Cr(VI) is given in Figure 4.

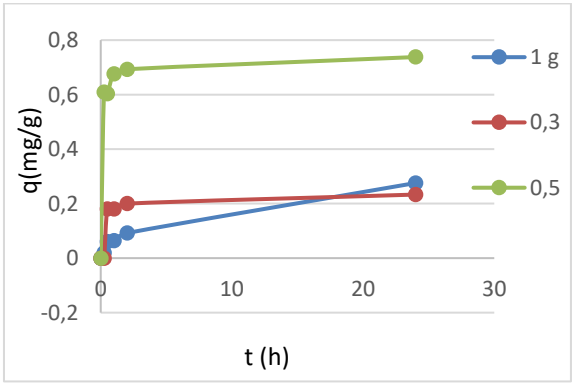

Figure 4. The effect of APK adsorbent amount on wastewater containing Cr(VI)

Şekil 4. APK adsorbent miktarının Cr(VI) içeren atıksu üzerine etkisi

3.2.3. *Effect of Initial Chromium Concentration*

In the adsorption experiments carried out in this study, it was observed that wastewater containing Cr(VI) with initial concentrations of 1, 2, 3, 4, 5, 10, and 20 ppm was adsorbed by APK at the end of the contact period of 5 hours, and the yield for the wastewater containing Cr(VI) was found to be at the beginning. The graph of the effect on the concentration is given in Figure 5.

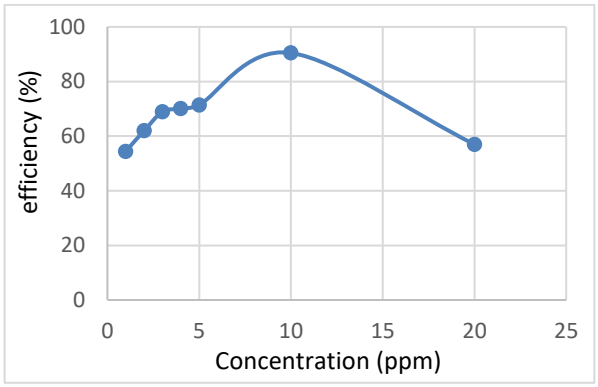

Figure 5. The effect of APK on the initial chromium concentration on wastewater containing Cr(VI)

Şekil 5. APK'nın Cr(VI) içeren atıksu için krom başlangıç üzerine etkisi

In the experiments, after 5 hours, and efficiency of 54.45% at a concentration of 1 ppm, 90.48% at a concentration of 10 ppm, and 57% at a concentration of 20 ppm was obtained.

3.2.4. *Effect of Contact Time*

According to Figures 3 and 4, q (adsorbent capacity) increased between 0-5 hours. Looking at the graphs, the constant q was fixed after the 5th hour. 5 hours was chosen as the optimum time for the adsorption studies.

3.2.5. *Adsorption Isotherms*

Langmuir and Freundlich Isotherm models were applied for the adsorption of Cr(VI)-containing wastewater with APK. Isotherm coefficients were calculated from the obtained data.

The coefficients of the isotherms are calculated using graphs and equations. The calculated coefficients are given in Table 1.

Table 1. Isotherm constants

Isotherm	Isotherm constants	Isotherm Values
Langmuir	q_m	0.0836
	b_a	28.993
	R^2	0.9486
Freundlich	n	0.892
	K_a	295.733
	R^2	0.9054

As a result of the studies, it has been seen that the Langmuir isotherm model is more suitable than the two isotherm models.

3.2.6. *Kinetic Studies*

First and second-order reaction kinetic models provide information about the kinetics of an adsorption system. In this study, first and second-order reaction rate kinetic models were

applied. Pseudo-first order and second-order kinetic models were examined for Cr(VI) removal of APK. The results showed that the R^2 value of the first-order kinetic model was 0.9021, and the R^2 value of the second-order kinetic value was 0.9582. The pseudo-second-order kinetic model was chosen for Cr(VI) removal. The fact that it was suitable for the pseudo-second-order kinetic model revealed that the reaction was chemical and that adsorbent bands could not be used again.

Table2. Kinetic constants

Kinetics	Parameters	Values
Pseudo-First- Order	K_1 (min^{-1})	1.828
	q_e ($mg.g^{-1}$)	5.876
	R^2	0.9021
Pseudo-Second-Order	K_2 ($g.mg^{-1}.min^{-1}$)	0.458
	q_e ($mg.g^{-1}$)	0.435
	R^2	0.9582

4. Conclusions and Recommendations

According to the experimental results we have done, the peach kernel was selected as an adsorbent in the ongoing experiments of the study and used in the ongoing experiments. In the study, pH 2, adsorbent amount of 0.5 g, and retention time of 5 hours were chosen as optimum conditions, and suitable results were obtained for Cr(VI) removal under these conditions.

It was observed that a removal capacity of 0.738 $mg.g^{-1}$ was obtained when 0.5 g APK was used as an adsorbent under optimum operating conditions. As a result of the application of the experimental data obtained as a result of the studies to the isotherm models, it has been seen that the Langmuir isotherm model is more suitable. In the reaction rate kinetic results for adsorption, our correlation coefficient R^2 was calculated as 0.9021 in the graph drawn for the first-order kinetic model and 0.9582 in the graph prepared for the pseudo-second-order kinetic model.

Based on the correlation coefficients in the evaluation made between both kinetic models, it was concluded that the pseudo-second-order kinetic model of the reaction rate kinetics was more appropriate in the Cr(VI) purification of APK. This result also concluded that the Cr(VI) removal of APK from the aqueous solution is a chemical reaction and that the reuse of the adsorbent after adsorption is not appropriate.

Today, the development of environmentally friendly technologies is an important research topic due to the reuse of wastes or alternative uses. It has been observed that APK can be considered as an alternative use in the removal of Cr(VI) from aqueous solutions. The results obtained have led to the conclusion that the use of APK as an adsorbent will provide toxic Cr(VI) removal in wastewater. This result can be considered as an advantage, as the cost is low economically and the reaction in the removal of adsorbed Cr(VI) is chemical, and there is no desorption of Cr(VI) into the water after the adsorption filler. Considering the APK calorific value after the adsorption process, it is possible to dispose of these facilities by contributing to the energy needs of the facilities where emission-controlled energy production and industrial production are carried out. As a result, APK can provide calorific value in units of suitable facilities. From another point of view, the recovery of Cr(VI) as a result of the need and economic evaluation as a result of the use of these facilities can be considered as a separate study.

Table 3 is showing comparison with other studies.

5. Acknowledge

The publication work obtained from this master's thesis is supported by Mersin University Scientific Research Projects Unit as a project numbered 2019-3-TP2-3712.

Table 3. Comparison with other studies

Adsorbent	Capacity ($mg.g^{-1}$)	Isotherm	Kinetic Model	References
Activated Peach Kernel	0.738	Langmuir	Pseudo-second-order	This study
Wheat residue	322.58	Freundlich	Pseudo-second-order	(Chen, Yue, Gao, & Xu, 2010)
Banana Peel	131.56	Langmuir and D-R	Pseudo-first-order	(Memon, and others 2009)
Rice husk	73.96	Langmuir	Pseudo-first-order	(Akhtar, Iqbal, Kausar, Bhanger, & Shaheen, 2010)

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