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Congo Red Removal From Aqueous Solutions by Adsorption on Peanut Hull

Fıstık Kabuğu Üzerinde Sulu Çözeltilerden Adsorpsiyon İle Kongo Kırmızının Uzaklaştırılması

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ABSTRACT

Congo red is one of the anionic diazo dyes and is widely used in the dying process for many industries. Industries such as textile, leather, paper, rubber, plastics and cosmetics are used the congo red dye. In this study, removal of congo red from aqueous solutions by adsorption on peanut hull was explored. The effects of several parameters like contact time, initial concentration, adsorbent amount, particle size and temperature were researched. A kinetic study was also performed. Adsorption rate increases up to 60mg/L initial concentration and then decreases. 60mg/L may be considered as an optimum initial concentration in the initial concentration range studied. The removal of congo red increases with increasing absorbent amount. The removal of congo red decreases with increasing adsorbent particle size and temperature as well. The adsorption equilibriums were reached at about 60min. Adsorption kinetics of congo red on the peanut hull followed by pseudo second - order kinetic model.

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ÖZET

Kongo kırmızısı anyonik diazo boyalarından biridir ve birçok endüstride boyama işleminde yaygın olarak kullanılmaktadır. Kongo kırmızı boyası tekstil, deri, kağıt, kauçuk, plastik ve kozmetik gibi sektörlerde kullanılmaktadır. Bu çalışmada fıstık kabuğu üzerinde sulu çözeltilerden Kongo kırmızısının uzaklaştırılması incelenmiştir. Temas süresi, başlangıç derişimi, adsorban miktarı, tane boyutu ve sıcaklık gibi parametrelerin etkisi araştırılmıştır. Adsorpsiyon hızı 60mg/L başlangıç derişimine kadar artmış, daha sonra derişimin artması ile azalmıştır. Artan adsorban miktarı ile kongo kırmızının uzaklaştırılması artmaktadır. Ayrıca adsorban partikül boyutu ve sıcaklığın artması ile kongo kırmızı uzaklaştırılması azalmaktadır. Adsorpsiyon yaklaşık 60 dakika sonra dengeye ulaşmaktadır. Fıstık kabuğu üzerinde kongo kırmızının adsorpsiyon kinetiği yalancı ikinci derece kinetik model izlemektedir

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1. INTRODUCTION

Congo red is one of the anionic diazo dyes and is widely used in the dying process for many industries. Industries such as textile, leather, paper, rubber, plastics and cosmetics are used the congo red dye. Congo red has a high molecular weight with aromatic complex structure. Due to complex structure, it has thermal, physicochemical and optical stability. It resists against oxidizing agents and biodegradation. Congo red has toxic, carcinogenic and mutagenic properties. These negative properties cause an adverse effect on the aquatic life, environment and public health [1- 4]. To protect the environment and human health, congo red must be removed from the water before releasing to aquatic environments.

Several treatment methods have been used to remove congo red from wastewater such as coagulation [5], photocatalysis [6], ozonation [7], sonication [8] and adsorption [2].

Among these methods, adsorption is a cheap, an effective and a simple treatment method with low energy consumption to remove dyes from aqueous solutions. Adsorption process involves the collection of molecules at the interface between two phases such as liquid–solid interface. The collected molecules at the interface are called adsorbate and the solids used for the adsorption process are defined as an adsorbent. In the adsorption process, adsorbent efficiency and cost are important. By using highly efficient and low-cost adsorbents, adsorption process becomes more economic, environmental friendly [2,9,10]. Adsorbents obtained from agriculture waste such as orange peel, coconut shell, rice husk, sugarcane bagasse and many more were used due to low-cost, non-toxic, easy to use and easily available [11,12].

Peanut hull is one of the agricultural waste and used as an adsorbent for the dyes. Peanut hull is easily available and has low-cost agricultural waste. In literature, peanut hull was used to remove different types of dyes such as crystal violet [12], anionic and cationic dyes [13,14], indisol orange RSN [15], indisol yellow BG [16], reactive black 5 [17], light green [18], congo red [19]. There are a few studies in literature has been reported on the adsorption of congo red on peanut hull.

Gurkan and Coruh [19] examined the effect of three different adsorbents on congo red removal. One of the adsorbents studied by the authors was peanut hull. In the study, shaking incubator at 150rpm was used. The contact time and initial concentration effect were investigated.

The aim of this study was to investigate the removal of congo red by adsorption using peanut hull as an adsorbent. In our study experiments were done using magnetic stirrer. In this study, the effects of contact time, an initial concentration of congo red solution, adsorbent amount, particle size and temperature were investigated. Kinetics of the reaction was also examined. Although there was a study using peanut shell as an adsorbent on congo red removal [19], in this study, equipment used for the adsorption experiments was different and the effect of various parameters were investigated.

2. MATERIALS AND METHODS

The peanut was supplied from a market and then peanut hull was separated from the peanut. The peanut hull was washed with distilled water to remove impurities like soil and dirt. After that, peanut hull dried in an oven until reaching a fixed weight. The dried peanut hull was cut into small pieces using a home type blender. A screening was performed to separate particles in varied sizes.

Congo red was obtained from Merck. Table 1 shows the properties of the congo red. A solution of the congo red was prepared by using distilled water. A stock solution was obtained by adding one gram of congo red dye per liter of distilled water. The congo red solutions with a known concentration of congo red were prepared using the stock solution and distilled water. The absorbance of the congo red solution for various concentrations was recorded using a UV/visible Spectrophotometer (Genesys10S, Thermo Scientific). Congo red dye wavelength, λ was found to be 490 nm. After recording the absorbance values of the congo red solutions at different concentrations, the standard curve was obtained.

Table 1. Properties of congo red dye

Congo Red	
Molecular structure	$C_{32}H_{22}N_6Na_2O_6S_2$
Molecular weight (g/mol)	696.68
Classification	Azo dye (N=N)
Melting point (°C)	>360
Bulk density (kg/m^3)	600-700
λ_{max} (nm)	497

In the study, magnetically stirrer (HSD-180, M-Tops), pH meter (C561, Consort) and centrifuge (CN 090, Nuve) were used.

For the adsorption study, the desired amount of the peanut hull as an adsorbent was put in 200 ml of congo red solutions and magnetically stirred continuously at 500 rpm. The stirring rate was constant in all the experiments.

The temperature of the congo red solutions was 25 °C except the experiments investigate the temperature effect. The pH of the solution was neutral about 7-7,5. Process variables like contact time, the initial concentration of congo red solution, amount of adsorbent, particle size and temperature were investigated. The samples were withdrawn from the reaction mixture and centrifuged at 4000 rpm for 10 minutes to remove adsorbent. The absorbance of the sample was evaluated to find the concentration of congo red.

Efficiency of the congo red removal, R was calculated as follows:

$$R, \% = [Co - Ct)/Co] * 100 \quad (1)$$

Equation 2 gives the adsorption capacity of the peanut hull as seen below:

$$q_e = \frac{(Co-Ce)V}{W} \quad (2)$$

where qe= adsorption capacity of the peanut hull at equilibrium (mg/g), Co= initial concentration of congo red, Ct= concentration of congo red at any time (mg/L), Ce= concentration of congo red at equilibrium (mg/L), V= volume of the congo red solution (L), and W= weight of the peanut hull used as an adsorbent (g).

3. RESULTS AND DISCUSSIONS

3.1. Effect of contact time

At the beginning of the study, contact time effect on the adsorption of congo red was investigated to find the equilibrium time. Figure 1 shows the contact time effect on the removal of congo red at 60 mg/L initial concentration, 0.2g/200ml absorbent amount and 25°C. The removal of congo red increased with time in one hour. The removal of congo red was nearly the same after one hour. It means the adsorption rate was rapid in one hour then reached equilibrium. For this reason other adsorption experiments were done for one hour.
At the beginning of the experiment active sites on the peanut hull surface are vacant for the binding of dye molecules, then the number of active sites decreased. After reaching equilibrium, binding sites on the peanut hull surface saturated and removal rate remained nearly constant. A similar result is obtained by Sadaf and Bhatti [15,16]. Zhang et al. [20] explored the removal of congo red onto soybean curd. Adsorption rate at the first 200 min was very rapid due to the interaction between the congo red molecules and the active adsorption sites on the adsorbent surface. After 200 min, adsorption rate decreases until the adsorption reaches equilibrium. Kataria and Garg [2] observed similar behavior for the adsorption of congo red. Adsorption rate increased at initial period due to the availability of a large number of the free surface active site onto the adsorbent. After some time adsorption of dye was reached equilibrium due to saturation of adsorbent surface. Nadi et al. [21] have studied the removal of reactive dyes using peanut shell. Reactive dye removal on a peanut shell increased rapidly with increasing contact time until 90 min.

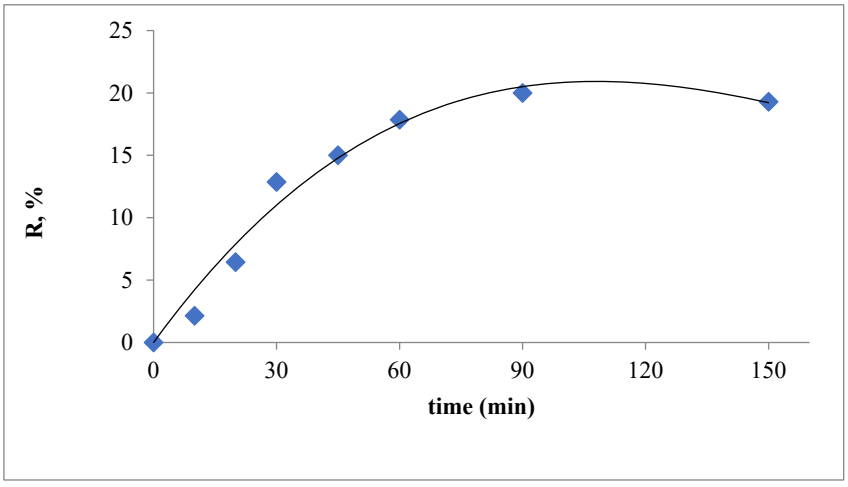

Figure 1. Effect of contact time on the removal of congo red (adsorbent amount:0.2g/200ml, particle size: 250 mic, congo red initial concentration: 60 mg/L, T: 25 °C)

3.2. Effect of congo red initial concentration

To investigate the initial concentration effect on congo red removal, four different initial concentrations, 40, 60, 80 and 100 mg/L were tested. Experiments were done with 200 ml aqueous solutions of congo red and 0.2g/200ml peanut hull amount at 25°C and 60 min reaction time. Figure 2 shows the results. As seen, the removal of congo red first increases with increasing initial concentration from 40 mg/L to 60 mg/L, then decreases with increasing initial concentration. At the end of the one-hour maximum congo red removal was obtained at 60 mg/L initial concentration.

In the adsorption process, the initial concentration of the dye is one of the important parameters. The high concentration provides a driving force to overcome mass transfer resistance between dye molecules and solid phase [12, 16, 22] . Samil et al. [23] studied initial concentrations of remazol orange RGB in the range of 10-150 mg/L using a peanut shell. Adsorption capacity and removal rate increased with increasing initial concentration. In literature similar results have been obtained for the removal of reactive dyes, indosol yellow BG and indisol orange RSN from aqueous solutions onto peanut husk [15,16, 21].

On the other hand, after optimum initial concentration adsorption rate decreases. At the studied adsorbent amount (0.2g/200ml), a number of adsorption sites limited to take congo red molecules. It means, there are more congo red molecules at higher concentrations for the same number of adsorption sites. Active sites saturated at high concentrations and congo red adsorption slowed down. Tanyıldızı [17] obtained similar results. According to the author, concentration effects adsorption rate inversely due to limited adsorption sites available for the uptake of dye. In literature, there are similar results for the adsorption of anionic and cationic dyes on peanut hull [13,14].

Tahir et al. [22] found that crystal violet adsorption on different types of adsorbents increased with increasing initial crystal violet concentration and reached its maximum at 150 mg/L initial concentration. After this initial concentration level, there was an insignificant change in the adsorption level due to saturation of binding sites of the adsorbent.

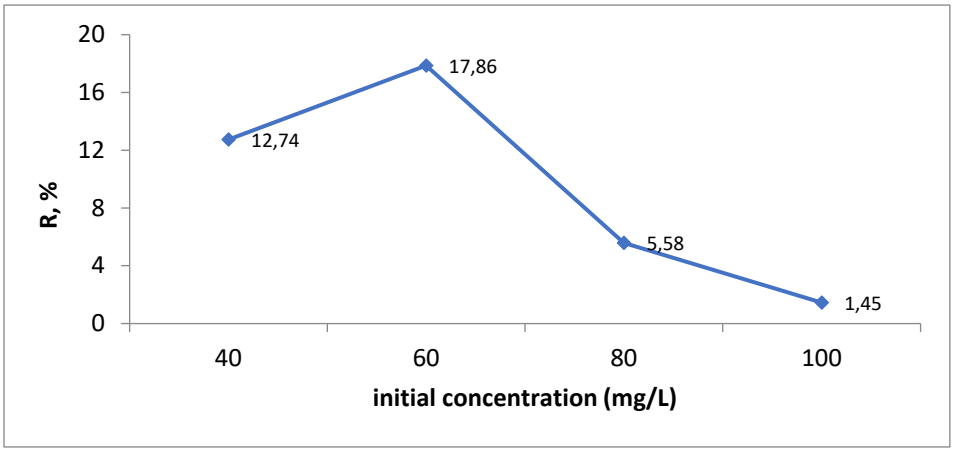

Figure 2. Effect of initial concentration on the removal of congo red (contact time 60 min, particle size: 250 mic, adsorbent amount:0.2g/200ml, T: 25°C)

3.3. Effect of adsorbent amount

In the present study, the effect of an adsorbent amount on congo red removal has been investigated. Experiments were done with a different adsorbent amount at 60 mg/L initial concentration and 25°C. Figure 3 presents the results of absorbent amount on the removal of congo red. The removal of congo red increases with increasing adsorbent amount in the range 0.2g/200ml – 1g/200ml.

Increase in removal rate with the adsorbent amount could be attributed to increased surface area and an increased number of active adsorption sites [2, 12-14]. A similar result was obtained for the adsorption of reactive black 5 on peanut hull [17]. In another study [20], adsorbent dose effect on the adsorption of congo red onto soybean curd was investigated. According to their results, the removal rate of congo red increases with increasing the adsorbent amount, due to the increased active adsorption sites at a higher adsorbent dose. Low et al. [12] have found that an increase in adsorbent dosage gave higher adsorption on crystal violet. Nadi et al. [21] studied the removal of reactive dyes from aqueous solutions by peanut shell powder. The removal efficiency of dyes increased with increasing adsorbent amount due to an increase in surface area.

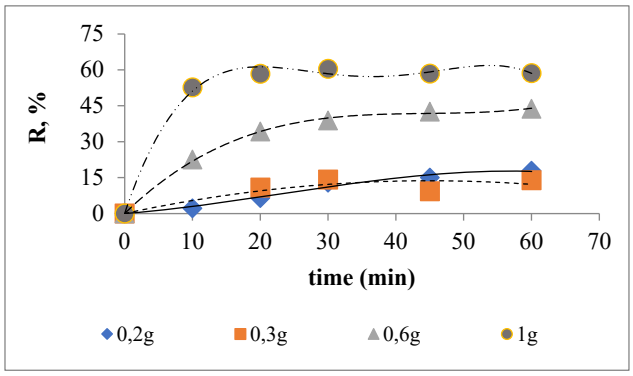

Figure 3. Effect of the adsorbent amount on the removal of congo red (contact time: 60 min, particle size: 250 mic, initial concentration: 60 mg/L, T: 25°

3.4. Effect of particle size

Particle size effect on the removal of congo red was studied with a particle size in the range of 180-350 mic using 0.6g/200ml peanut hull amount and 60 mg/L initial concentration at 25°C and 60 min contact time. As seen in the previous section, maximum congo red removal was obtained at 1g/200ml adsorbent amount. To investigate particle size effect, adsorbent amount was choosen as 0.6g/200ml instead of 1g/200ml due to difficulty in seperation. Figure 4 represents the results. As shown from Figure 4, congo red removal increases with decreasing the adsorbent particle size. The surface area of the adsorbent increases with decreasing particle size. For this reason, the removal rate increases. Gong et al. [13,14] obtained similar results. Although the maximum removal rate was obtained at the particle size 180 mic, other experiments were done with 250 mic due to the easiness of liquid and solid separation.

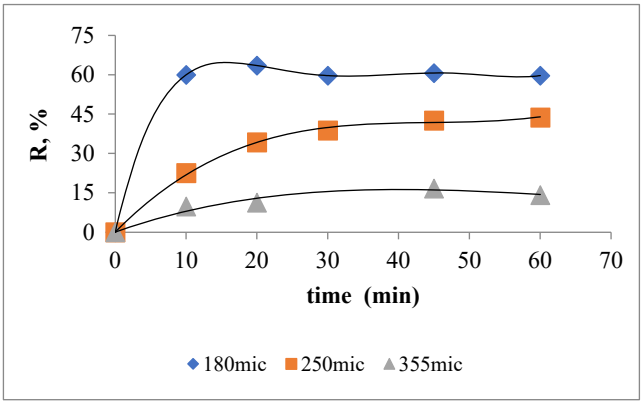

Figure 4. Effect of particle size on the removal of congo red (contact time: 60 min, adsorbent amount: 0.6g/200ml, initial concentration: 60 mg/L, T: 25°C)

3.5. Effect of temperature

Temperature effect on the removal of congo red was studied at 25,35, 50°C using a congo red solution having 60 mg/L concentration and 0.6g/200ml peanut amount for 60 min. As shown in Figure 5, the removal rate decreases with increasing temperature. This result indicated that the adsorption process was exothermic. Increase in temperature may cause decrease in the adsorptive forces between the dye molecules and the active sites on the adsorbent surface [9]. A similar result was observed by Sadaf and Bhatti [15,16] and Zhao et al.[18]. In another study, Kataria and Garg [2] said that the solubility of anionic dyes increases with increasing temperature. Congo red removal using ZnO nanoparticles as an absorbent decrease with increasing temperature. According to this result, the adsorption process is exothermic. It may be based upon to desorption of dye molecules by the internal heat energy of adsorbent at a higher temperature.

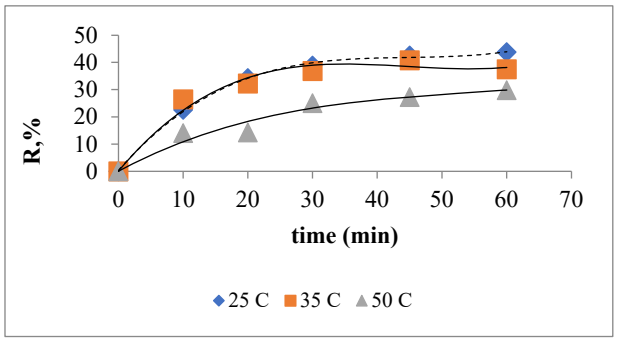

Figure 5. Effect of temperature on the removal of congo red (contact time: 60 min, adsorbent amount: 0.6g/200ml, particle size: 250 mic, initial concentration: 60 mg/L)

3.6. Kinetic models for congo red adsorption on peanut hull

The kinetics of the congo red adsorption on peanut hull was considered using the pseudo first - order model and pseudo second - order model. Kinetic constants for each model were found for three different temperatures.

The pseudo first - order kinetic model is expressed as a linear form by using Eq. 3.

$$\ln [q_e - q_t] = \ln q_e - k_1 t \quad (3)$$

where q_e adsorbed congo red amount at equilibrium (mg/g), q_t adsorbed congo red amount at any time (mg/g), and k_1 (min^{-1}) the rate constant of pseudo first - order adsorption. If $\ln[q_e-q_t]$ is plotted versus time a straight line would be achieved with the slope being k_1. q_e value is found using intercept value [11,17].

Equation 4 gives the linear form of the pseudo second order kinetic model

$$[t/q_t] = [1/(k_2 q_e^2)] + [t/q_e] \quad (4)$$

where k_2 (g/mgmin) rate constant of pseudo second order adsorption. If $[t/q_t]$ is plotted versus time, obtained straight line gives the adsorption parameters of the pseudo second - order kinetic model. Adsorbed congo red amount at equilibrium (q_e) was found from the slope of the straight line and rate constant (k_2) was found from the intercept of the straight line [11,17].
The suitability of these kinetic models is determined by correlation coefficients (R2). In addition to this, the conformity of experimental and calculated qe values is important to specify the model. If the correlation coefficient (R2) is high and the difference between experimental values of qe and calculated values of qe is small, it can be said that the model is best.
The variation of ln[qe − qt] with time and the variation of [t/qt] with time at various temperatures are given in Figures 6 and 7 respectively.

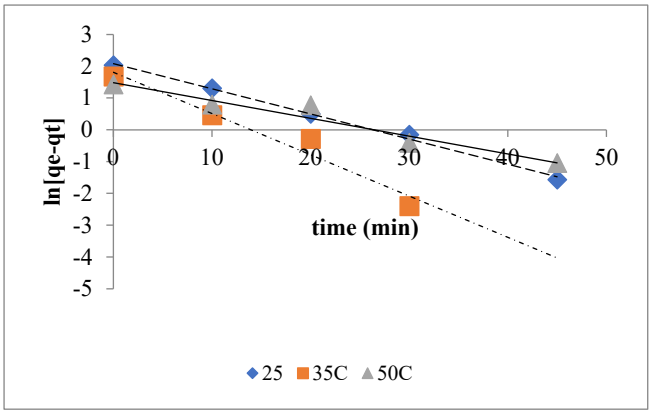

Figure 6. The variation of $\ln[q_e - q_t]$ with time at different temperatures (60 mg/L initial concentration, 0.6g/200ml absorbent amount and 60 min contact time)

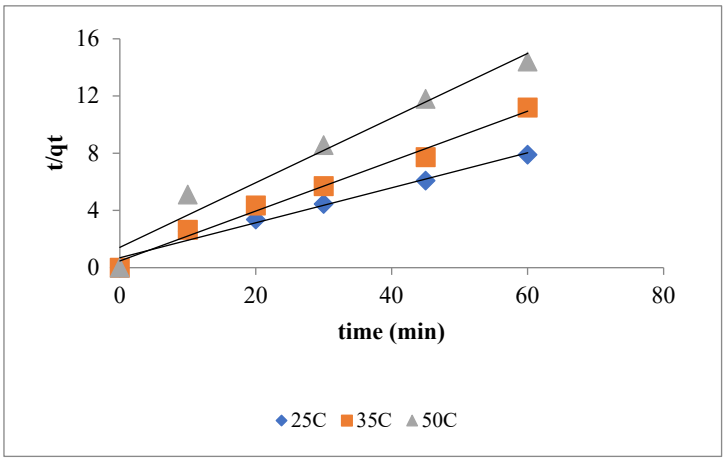

Figure 7. The variation of [t/q_t] with time at different temperatures (60 mg/L initial concentration, 0.6g/200ml absorbent amount and 60 min contact time)

Table 2 presents the values of kinetic rate constants and correlation coefficient (R2) for each model. Experimental and calculated values of the adsorbed congo red at equilibrium also given in Table 2. The correlation coefficient (R2) values of the pseudo second - order model were slightly greater than that of the pseudo first - order model. Furthermore, the experimental qe values and calculated qe values are closer to each other. Therefore, it can be said that pseudo second - order kinetic model for the adsorption of congo red on peanut hull is more favorable than the pseudo first - order kinetic model in studied experimental conditions. This result may happen due to chemisorption of congo red dye molecules on the peanut hull [11,17]. In literature similar results have been obtained for adsorption of congo red using various adsorbents [11, 20, 24].

Table 2. Parameters of different kinetic models for congo red adsorption on peanut hull

T (°C)	$q_{exp.}$ (mg/g)	Pseudo first order model			Pseudo second order model			Intra particle diffusion		
		k_1 (min^{-1})	$q_{e,calc}$ (mg/g)	R^2	k_2 (g/(mgmin)	$q_{e,calc}$ (mg/g)	R^2	k_i	C	R^2
25	7.6	0.0792	8.03	0.996	0.022	8.18	0.975	0.214	0,5749	0.950
35	5.36	0.1298	6.11	0.958	0.066	5.72	0.987	0,7262	0.8006	0.877
50	4.16	0.056	4.4	0.942	0.036	4.42	0.965	0,5531	0.0308	0.958

4. CONCLUSIONS

In this study, removal of congo red from aqueous solutions by adsorption on peanut hull was explored. The effects of several parameters like contact time, initial concentration, adsorbent amount, adsorbent particle size and temperature were investigated. A kinetic study was also performed.

Adsorption rate increases up to 60 mg/L initial concentration and then decreases. 60 mg/L may be considered as an optimum initial concentration in the initial concentration range studied. The removal of congo red increases with increasing absorbent amount. The removal of congo red decreases with increasing particle size and temperature as well. The adsorption equilibrium was reached at about one hour. The adsorption of congo red followed the pseudo second - order kinetics. This study showed that peanut hull as an adsorbent could effectively remove congo red from aqueous solution

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