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Research Article

INVESTIGATION THE REMOVAL OF MALACHITE GREEN DYE FROM AQUEOUS SOLUTION BY USING SESAME SHELL

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Abstract

In this study, the removal of Malachite Green (MG) dye from the aqueous solution by the adsorption method onto Sesame Shell (SS) was investigated. The effects of solution pH, the initial concentration of the dye, adsorbent amount and contact time on the adsorption process were determined. The adsorption kinetic was modeled using first order and pseudo second order kinetic models. Langmuir and Freundlich isotherms were applied in this study. All of the experiments were carried at the room temperature (25°C) and at solution volume of 30 mL. The optimum results had been noticed at the following parameters pH 8, initial concentration of 20 mg/L, the adsorbent amount of 0.3 g and contact time was 40 minutes. The adsorption of MG onto SS was found to be more suitable for the pseudo second order kinetic model with correlation factor of R^2=0.9998. The result was fitted with Langmuir isotherm (R^2=0.9997). The adsorbent capacity was determined to be 1.58 mg/g, the removal efficiency of MG onto SS was 93.45%. In this study, the SS was used as an adsorbent for MG removal and based upon the obtained results it is found to be an effective choice. On the other hand, it was concluded that the calorific value of 4122 Cal/g determined for the contaminated adsorbent was an attractive waste for incineration plants for the disposal of the contaminated adsorbent.

SULU ÇÖZELTİDEN SUSAM KABUĞU KULLANILARAK MALAKİT YEŞİLİ BOYASININ GİDERİMİNİN ARAŞTIRILMASI

Anahtar Kelimeler
Adsorpsiyon,
Susam kabuğu,
Malakit yeşili,
Boya,
Adsorban, Tarımsal atık.

Özet

Bu çalışmada, Susam Kabuğu (SS) üzerine sulu çözeltiden Malachite Green (MG) boyasının adsorpsiyon yöntemi ile uzaklaştırılması araştırılmıştır. Boya çözeltisinin pH, başlangıç derişimi, adsorban miktarı ve temas süresinin adsorpsiyon işlemine etkileri belirlenmiştir. Adsorpsiyon kinetiği modelini belirlemek için birinci ve yalancı ikinci mertebeden kinetik modeller kullanılmıştır. Çalışma için Langmuir ve Freundlich izoterm modelleri uygulanmıştır. Tüm deneysel çalışmalar oda sıcaklığında (25°C) ve 30 mL çözelti hacminde gerçekleştirilmiştir. Optimum sonuçlar; pH 8, başlangıç derişimi 20 mg/L, adsorban miktarı 0.3 g ve temas süresi 40 dakika olarak tespit edilmiştir. MG'nin SS üzerine adsorpsiyonunun, korelasyon faktörü R^2=0.9998 olan yalancı ikinci derece kinetik model için daha

uygun olduğu ve Langmuir izotermine (R^2=0.9997) uygun olduğu tespit edilmiştir. Adsorban kapasitesi 1.58 mg/g olarak belirlenmiş olup MG'nin SS üzerine giderim verimi %93.45 olarak hesaplanmıştır. Bu çalışmada elde edilen sonuçlara göre SS, MG giderimi için adsorban olarak kullanımı için etkili bir olduğu bulunmuştur. Öte yandan kirletilmiş adsorban için belirlenen 4122 Cal/g kalorifik değeri, kirlenen adsorbanın nihai bertarafı için yakma tesislerinde cazip yakıt olabileceği sonucuna varılmıştır.

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1. INTRODUCTION

Dyes are chemical compounds that can attach themselves to surfaces or fabrics to give color [1]. These dyes are used in many industries, textiles, leather, cosmetics, paper, printing, plastics, pharmacy, food, pisciculture, etc., in color products that produce large quantities of wastewater with high and organic content. The development of the rapidly growing textile industry in the world in recent years has led to an increase of textile wastewater. Only two thirds of the total wastewater containing dyestuffs is the textile industry [2]. Various physical, biological, and chemical techniques may be employed for the decolorization of dyes found in wastewater but, each method has technical and economic limitations [3, 4, 5, 6]. Many physicochemical decolorization methods are not suitable because they are expensive, have restricted usage areas, interfere with other wastewater components, or cause wastes that require retreatment. The biologic treatment method is an alternative to the physicochemical methods which is relatively inexpensive and may be preferred for decolorization [7, 8]. Textile wastewater is discharged without sufficient purification, affecting the chemical structure of the receiving water source, disrupts the aesthetic appearance, prevents the passage of sunlight into the water environment, and therefore reduces photosynthetic action. All of these have negative effects on the aquatic ecosystem [9, 10]. The Water Pollution Control Regulation in Turkey in Table 10, the color parameter according to the discharge standard is 280- 260 (Pt-Co) [11]. Since approximately 10-15% of the dyes used are discharged into the water environment, disposal of dyes is a serious global issue [12]. Malachite green (MG) is a triphenylmethane cationic dye [13]. MG has been extensively

used in the dyeing of leather, silk, paper, textile industries, bactericide, fungicide, and a parasiticide [14, 15]. Despite its many uses, MG is highly toxic to aquatic fauna, flora and humans due to its mutagenicity, carcinogenicity, and genotoxicity. Therefore, the removal of wastewater from MG dye is very important [13-16]. There are many dye removal methods from wastewater. Some of these are membrane filtration [17], adsorption [18], ozonation [19], oxidation [20], coagulation-flocculation [21], and photodegradation [22] methods. A few of them are effective, although they involve the use of concentrated chemicals that bioaccumulate or require expensive equipment for environmental improvement [23]. Adsorption is the most preferred advanced treatment method due to its efficiency, simplicity and insensitivity to the toxicity of pollutants [24]. Adsorption is a method of low cost, environmentally friendly, high regeneration capacity and used in the efficient treatment of wastewater [25]. Adsorption is the removal of particles that use the transfer of mass from the liquid phase to the solid phase [26, 27]. Activated carbon is one of the most popular adsorbents due to its high adsorption capacity. However, adsorbents are expensive and difficult to recycle. In this context, the use of natural and low-cost adsorbents in adsorption, especially the use of agricultural wastes, is an effective and economical method for cleaning dyes from textile wastewater [22]. Sesame, *Sesamum indicum L.* is an agricultural production herb that provides seeds for confectionery, edible oil, tahini (paste), cake and flour since ancient times [28, 29]. Sesame plant grows in tropical and temperate climates. The need for water is low, it can give good yield at high temperatures and the grain quality is high [23]. Sesame seeds contain 50-60% oil, 30-35% protein and 1.3% calcium and minerals in terms of their content [30]. According to the Turkey Statistical Institute data of 2020 last two years (2019-2020) 16 893 tons respectively 18 648 tons of sesame production it has been realized [31]. The seeds of the sesame plant which can be used in many sectors such as the food and pharmaceutical industry are usually burned or discarded after the interior is removed. As a result, the remaining sesame shell forms an agricultural waste [32, 33]. In this study, the usability of SS, which is an agricultural waste, was investigated in the wastewater treatment that contains MG dyestuff as an adsorbent.

2. MATERIALS AND METHODS

2.1. Adsorbent Preparation

The SS supplied from Gaziantep province was kept at room temperature (25 ˚C). It was washed with tap water first and then with pure water. After that, it was dried at scientific HERAEUS oven in 105 ˚C for 24 hours. The dried adsorbent was grinded with ISOLAB blender and 60 mesh sieves for later use. It's shown in Figure 1.

Figure 1. Preparation of Adsorbent

2.2. Dye Preparation

The cationic MG dye prepared 1000 ppm stock solution from SIGMA-ALDRICH. The MG dye has a chemical formula of $C_{23}H_{25}CLN_2$ (Figure 2) and its molecular weight is 346.9 g/mol. In experiments, 0.1 M NaOH and 0.1 M HCl were used to adjust the pH. The study was carried out on the values obtained by determining the highest adsorbing capacity of the dyestuff in the aqueous solution of Sesame Shell.

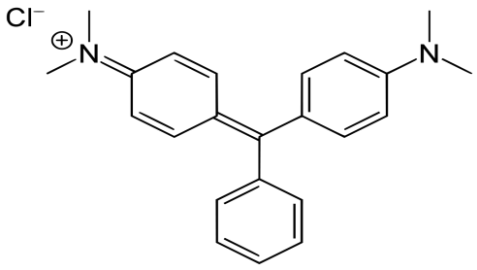

Figure 2. Chemical Formula of MG dye

2.3. Adsorbent Characterization

In adsorption experiments, ISOLAB precision scales were used for weighing; STUART BIOLAB SSL1 to mix solutions at 150 rpm, ISOLAB-WATERPROOF IP57 pH meter was used to determine the acid-base degree. In determining the color change in the experiments, UV spectrophotometer (HACK DR-3900) with a wavelength of 617 nm was used. The s synthesis composites before and after MG adsorption were recorded using Fourier Transform Infrared Spectroscopy (FTIR) [34, 35]. Scanning Electron Microscope (SEM) was used in the morphological investigation of composite structures [34].

2.4. Adsorption Experiments

In experiments, pH (2, 4, 6, 8, 10), initial concentration of dye (5, 10, 15, 20, 25 mg/L), adsorbent quantity (0.1, 0.3, 0.6, 0.9, 1.2 g) and time (5, 10, 20, 40, 80 min) were optimized. In all of the experiments, 30 mL solution was used. The solutions were placed in 100 mL flasks in case of overflow during shaking. Samples were then taken from each flask and centrifuged (HETTICH MICRO 22R) at 6000 rpm for 5 minutes. After centrifugation, samples were taken from the rinse phase at the top of each sample. After the experiment, adsorbent capacity (q_e) (mg/g) and efficiency (%) were calculated using Equation 1 and Equation 2 [36].

$$q_e = \frac{(C_i - C_f) \times V}{m} \qquad \textbf{Equation 1}$$

$$Efficiency = \frac{(C_i - C_S)}{C_i} x\ 100 \qquad \textbf{Equation 2}$$

In the equations, C_i is the initial concentration (mg/L) of the MG dye; C_f is the MG final concentration (mg/L); V solution volume (L); m indicates the adsorbent mass (g).

2.5. Adsorption Isotherms

In the adsorption process, Langmuir and Freundlich isotherms are commonly used to describe the relationship between adsorbent and adsorbent. Langmuir isotherm indicates monolayer adsorption and

homogeneous regions in the adsorbent (Equation 3) [2]. Freundlich isotherm defines multi-layer and heterogeneous systems (Equation 4) [35].

$$\frac{Ce}{q_e} = \frac{1}{K_L} + \left(\frac{a_L}{K_L}\right) C_e \qquad \textbf{Equation 3}$$

$$\mathrm{Log}(q_e) = \log(K_f) + \left(\frac{1}{n}\right) \log(C_e) \qquad \textbf{Equation 4}$$

In the equations, C_e is the concentration of adsorbate at the equilibrium (mg/L). K_L and a_L are Langmuir isotherm constant related to the affinity of the binding sites and energy of adsorption (L/mg). q_m is monolayer adsorption capacity (mg/g). K_f is the Freundlich adsorption capacity parameter (mg/g) (L/mg). 1/n is the intensity parameter.

2.6. Adsorption Kinetics

Adsorption kinetics are used to express the behavior of the reaction that occurs during adsorption experiments. In this study, pseudo first order (Equation 5) and pseudo second order (Equation 6) kinetic models were applied [37].

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

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

q_e: the amount of dye adsorbed at equilibrium (mg/g)

q_t,: amount of dye adsorbed at any time (mg/g)

K_1: rate constant of adsorption (the pseudo first order) (1/min)

K_2: rate constant of adsorption (the pseudo second order) (g/mg.h)

3. CONCLUSION AND DISCUSSION

3.1 Characterization of Adsorbent

In the study, SEM image and FTIR were used to examine the functional group analysis of the SS used as an adsorbent.

3.1.1. Scanning Electron Microscope Review

In the study, SEM examination was carried out to see the changes in the surface morphology of the SS used as an adsorbent before the adsorption process and after the adsorption process took place, that is, after the dyestuff in the aqueous solution was adsorbed by the SS. The SEM image taken before the adsorption process is given in Figure 3 and the SEM image after the procedure is given in Figure 4.

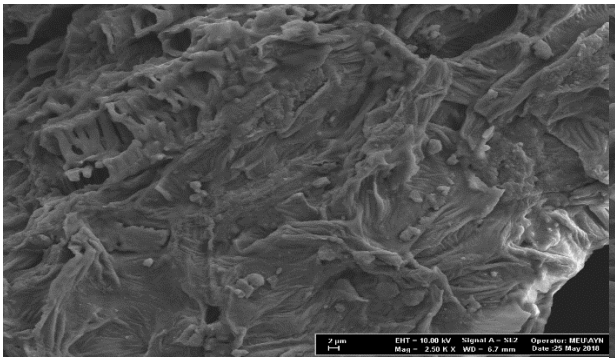

Figure 3. SS surface before adsorption process (250X)

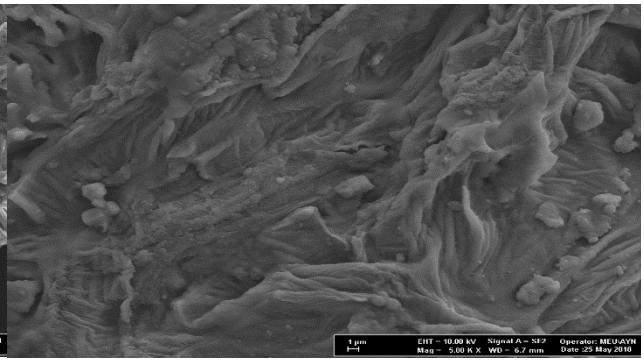

Figure 4. SS surface after adsorption process (500X)

When the results of SEM analysis are examined, the pores that can be seen in Figure 3 before the adsorption and the indented-protruding heights on their surface; As can be seen easily in Figure 4, it is seen that the surface is covered by the dyestuff after the adsorption process.

3.1.2. Fourier Transform Infrared Spectroscopy Analysis

FTIR device used in the study; It is a device used to determine the characterizations of organic or inorganic substances. Absorption peaks occur depending on the frequency that occurs as a result of the

vibrations of the functional groups that make up the substance. Each substance has its own spectrum [38]. In other words, if the examined item has formed a bond with any other substance, the peak values that occur depending on the spectrum due to the vibration spectrum of the new bond will change. FTIR analysis of raw (undyed) and dyed version of SS is shown in Figure 5.

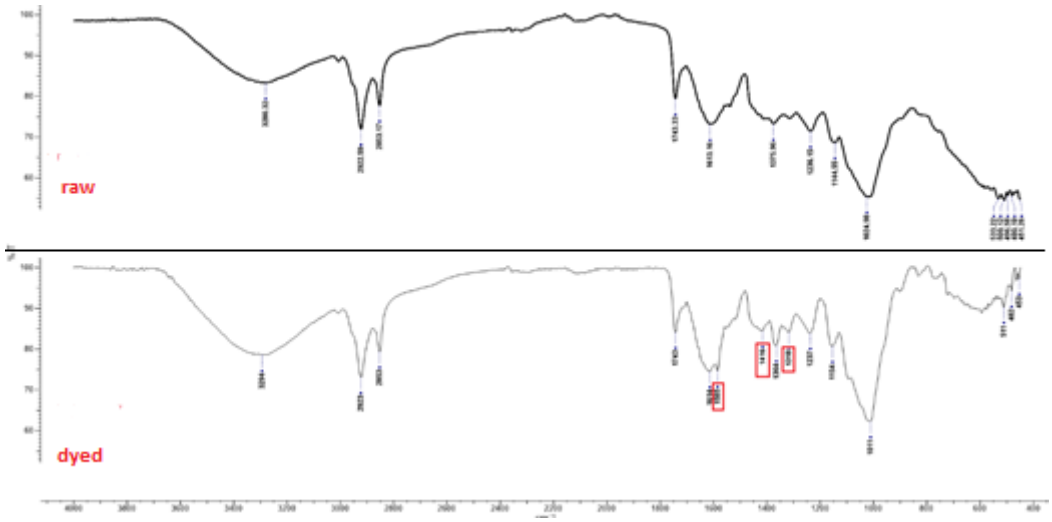

Figure 5. Raw and dyed FTIR analysis of SS

The adsorption values corresponding to each frequency region are different. These values may give different peaks according to different bond structures. As seen in Figure 5, when the FTIR analysis of SS in raw and dyed form is examined and compared, it shows that new bond structures are established during the adsorption of 1318 cm^{-1}, 1416 cm^{-1} and 1585 cm^{-1} values shown in red areas. The first peak of 1318 cm^{-1} refers to the C-N bond, peak to 1416 cm^{-1} refers to the C-H bond and a peak of 1585 cm^{-1} refers to the N-H bond.

3.2. Adsorption Optimization

3.2.1 pH Optimization

In the adsorption process where SS is used as an adsorbent; 30 mL aqueous solutions with concentrations of 100 mg/L was determined at pH 2, 6, 4, 5, 8 and 10 in order to determine the optimum working pH of the SS in aqueous solution containing dye is set to be. In the study where all other parameters were kept constant, 0.3 g of adsorbent was added to each flask. The samples taken

into the shaker were mixed for 24 h and then centrifuged at 6000 rpm for 5 min and measuring were made on the spectrophotometer. The values found as a result of the measurements are given in Figure 6.

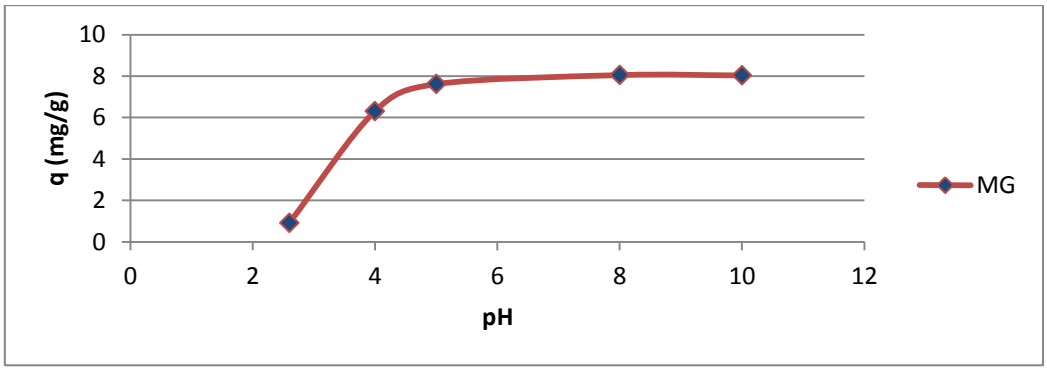

Figure 6. The effect of pH on SS (C_i: 100 mg/L, T: 25 ˚C, W: 0.3 g, t: 24 h)

As can be seen in Figure 6, as a result of the study carried out in order to determine the optimum working pH of the SS by keeping the other parameters constant, it was determined that the SS reached the highest adsorption capacity at pH 8. For this reason, the pH of the solution was preferred as 8 in the subsequent optimization studies. The pH of the aqueous solution containing dye used in the study was found to be 5, but as can be seen in the graph given in Figure 6, the highest adsorption capacity for SS was achieved with 8.048 mg/g at pH 8.

3.2.2. Initial Dyestuff Concentration Optimization

In order to allow the SS to adsorption the dyestuff in the aqueous solution at the highest adsorption capacity, 30 ml of aqueous solutions with concentrations of 5, 10, 15, 20, 25 mg/L have been studied. In the study in which all other parameters were kept constant, after centrifugation at 6000 rpm for 5 min at the end of 24 h, the spectrophotometer measuring was made. The results of the readings are given in Figure 7 below.

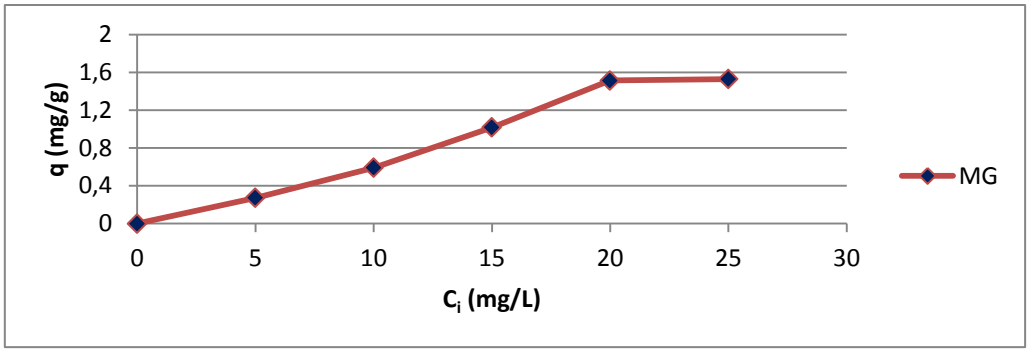

Figure 7. Effect of initial dyestuff concentration on SS (pH 8, W: 0.3 g, t: 24 h)

As can be seen in the graphic in Figure 7, when the effect of the initial dyestuff concentration on the SS is kept, other parameters are fixed, it is determined that the SS reaches the highest adsorption capacity (1.514 mg/g) at 20 mg/L initial dyestuff concentration. As can be seen in the graphic, it was determined that the adsorption capacity of SS was balanced after the initial concentration of 20 mg/L, that is, the absorbent reached saturation. For this reason, the optimum initial dyestuff concentration was preferred as 20 mg/L in next studies.

3.2.3. Optimization of Adsorbent Quantity

In order to find the adsorption capacity depending on the amount of SS used as an adsorbent under the initial concentration and to determine the removal efficiency depending on the adsorbent quantity of the dyestuff in the aqueous solution, 0.1, 0.3, 0.6, 0.9 and 1.2 g of adsorbent were added. Then, the samples taken on the shaker were mixed and then centrifuged at 6000 rpm for 5 min and the samples taken from each were measure on the spectrophotometer. The values found for each sample taken are given in Figure 8 and Figure 9 below.

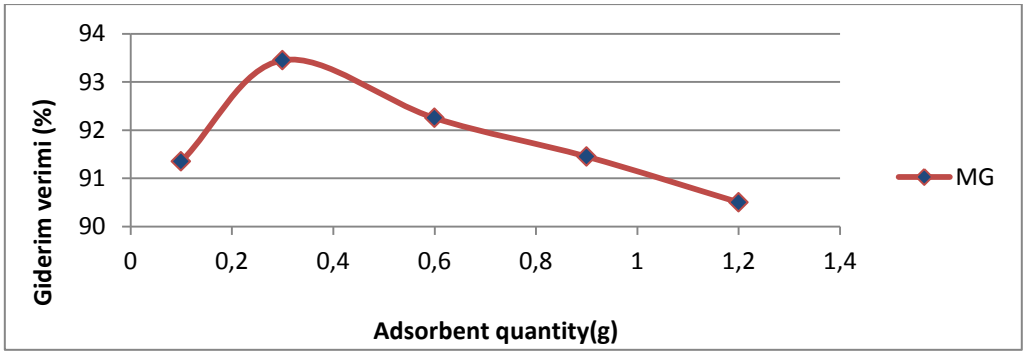

Figure 8. The effect of adsorbent amount on MG removal efficiency (T: 25 ˚C, V: 30 ml)

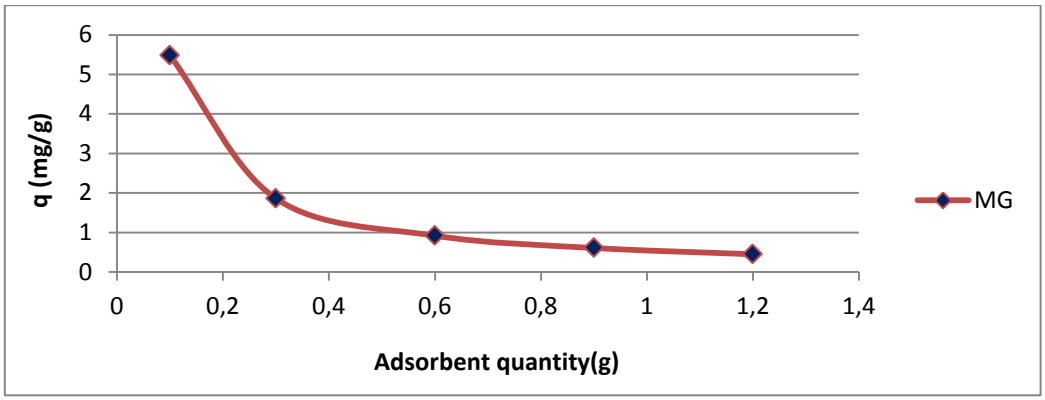

Figure 9. Effect of adsorbent quantity on MG (T: 25 ˚C, V: 30 mL)

When we look at Figure 8, the effect of different adsorbent quantity on the adsorption capacity was examined and as can be seen in the chart given in Figure 10, the highest adsorption capacity was reached in the quantity of 0.1 g adsorbent put into solution. As seen in the graph, it is seen that the adsorption capacity decreases due to the increasing quantity of adsorbent in the solution. As a result of the researches, it was found that as the amount of adsorbent in the aqueous solution increases, the quantity of dye that can adsorb in the solution per unit adsorbent decreases.

As it can be seen in the graph given in Figure 9, the effect of different adsorbent quantity on the removal efficiency of the dyestuff in the aqueous solution was examined and it was found that the highest removal was found as 0.3 g with the 93.45% removal rate. For this reason, 0.3 g was preferred as the optimum adsorbent quantity in the study.

3.2.4 Time Optimization

In order to determine the optimum time during which the adsorption capacity reaches equilibrium; other parameters are kept constant, pH 8; Solutions of 100 mL, 5, 10, 15 and 20 mg/L concentration were prepared. The solutions with 1 g of adsorbent were added to the shaker at the same time and the samples were taken at 5, 10, 20, 40, and 80 minutes from each solution. After centrifuged at 6000 rpm for 5 min and measure on the spectrophotometer. The values found for the samples taken at the times determined for each solution are given in Figure 10 below.

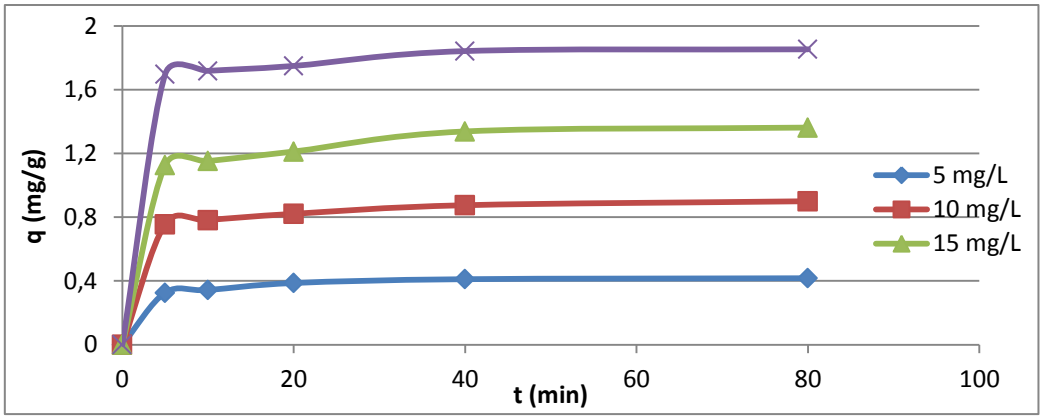

Figure 10. Effect of contact time on SS (pH 8, W: 1 g, V: 100 mL, T: 25 ˚C)

As can be seen in the graphic given in Figure 10, adsorption capacity has reached the balance by saturation after 40 minutes. Therefore, 40 minutes are preferred as the optimum contact time. As a result of our studies in samples with concentrations of 5, 10, and 15 mg/L, which we applied except for the optimum starting concentration of 20 mg/L, as shown in the graph given in Figure 10, although different concentrations were used, the optimum contact time did not change and the balance was reached in the same time.

3.3 Adsorption Isotherms

Langmuir and Freundlich Isotherm models for SS used as an adsorbent with MG dyestuff in aqueous solutions were investigated. The Langmuir Isotherm was created by drawing the change graph of C_e/q_e calculated against the balance against C_e (Figure 11). The Freundlich Isotherm model was created in the change graph of the $\log q_e$ calculated in the state of equilibrium against the $\log C_e$ (Figure 12). The isotherm coefficients calculated using the isotherm charts given in Figure 11 and Figure 12 are given in Table 1.

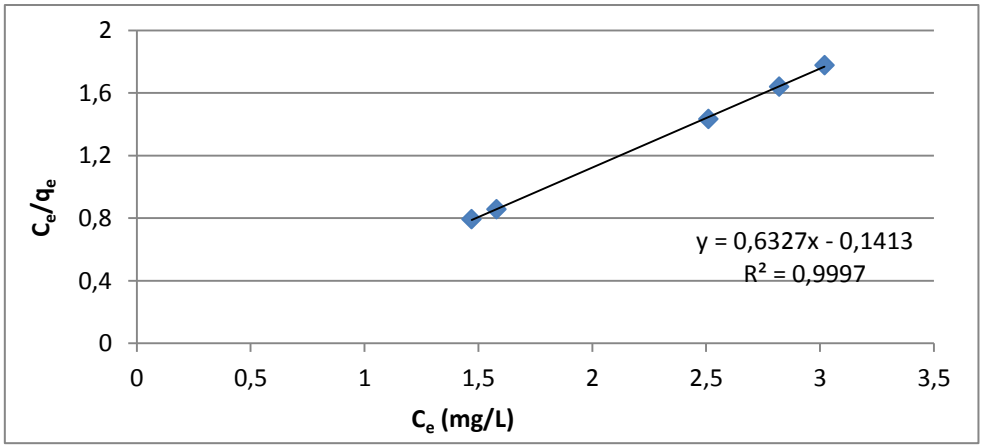

Figure 11. Langmuir Isotherm chart

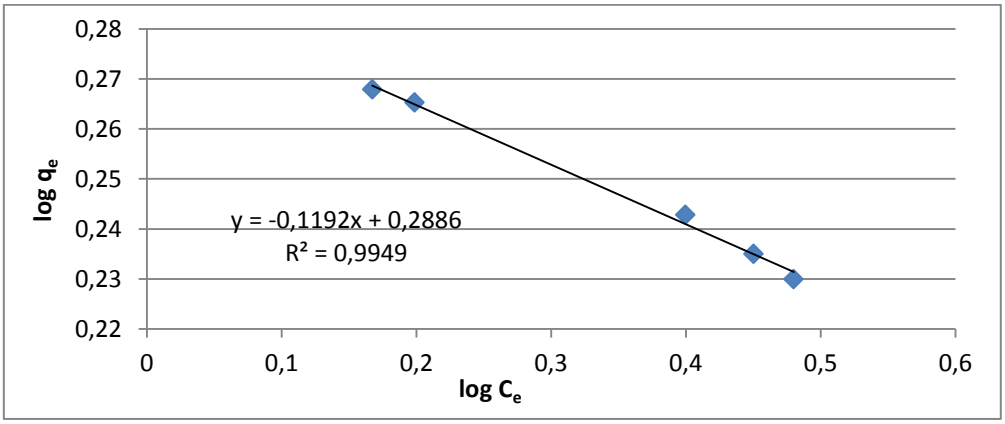

Figure 12. Freundlich Isotherm chart

Table 1. Langmuir and Freundlich Isotherm constants

	Isotherm Constants
Langmuir Isotherm	K_L= - 7.07714 a_L= - 4.47771 R^2= 0.9997
Freundlich Isotherm	K_F= 1.9435 n= - 8.38926 R^2= 0.9949

When Figure 11 and Figure 12 are examined, the number of regression of the Langmuir Isotherm model (R^2=0.9997) is higher than the number of regression of the Freundlich Isotherm model (R^2=0.9949). Therefore, the Langmuir Isotherm model was considered as the best isotherm model.

3.4. Adsorption Kinetics

In this study, the pseudo-first-order kinetic model and pseudo-second-order kinetic model calculations were made to determine the adsorption kinetics. Time (min) graph was drawn against log (q_e-q_t) to find the pseudo-first-order kinetic finding in order to find the change of adsorption capacity change rate over time (Figure 13). In order to calculate the rate of change of adsorption capacity over time, t graph was drawn against t/q_t was plotted to find pseudo-second-order kinetics (Figure 14). The coefficients calculated for the pseudo-second-order kinetic model are given in Table 2.

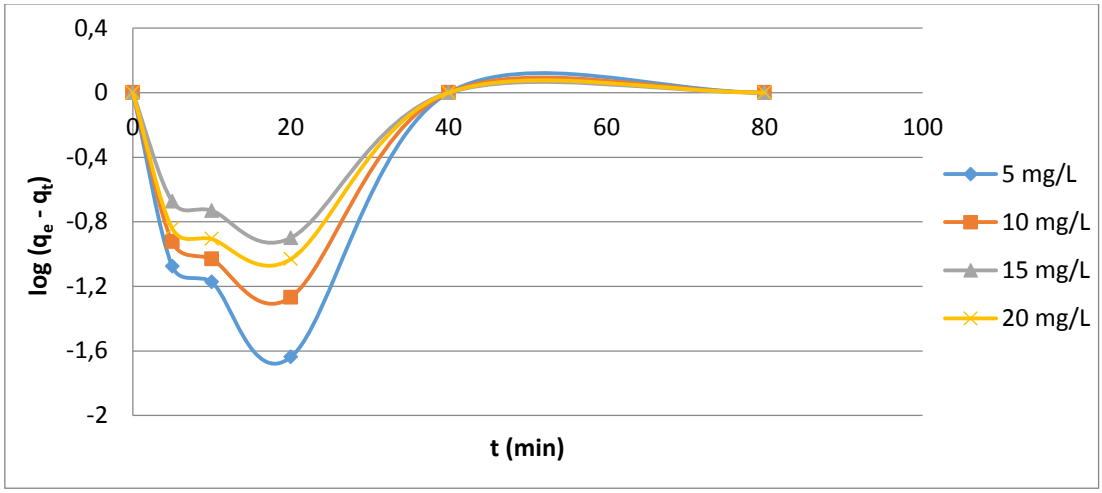

Figure 13. Pseudo-first-order kinetic model (MG-SS)

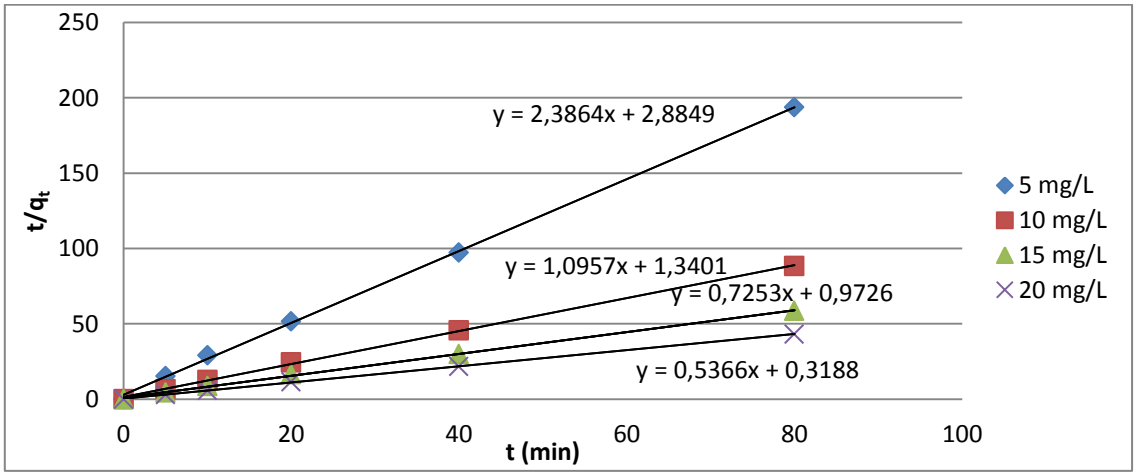

Figure 14. Pseudo Second order kinetic model (MG-SS)

Table 2. Coefficients calculated for pseudo second order kinetic model

Concentration (mg/L)	**q_e (mg/g) (experimental)**	**q_e (mg/g) (calculated)**	**k_2 g/mg.dk**	**R^2**
5	0.41	0.4190	1.9740	0.9994
10	0.875	0.9126	0.8958	0.9993
15	1.338	1.3787	0.5408	0.999
20	1.842	1.8635	0.9031	0.9998

Considering the R^2 (regression) numbers given in Table 2, it is seen that the pseudo-second-order kinetic model is suitable for use in the adsorption of MG onto the SS.

4. Previous Studies

In this part, the comparison of this study with low-cost adsorbents previously is shown in Table 3.

Table 3. Comparison of other adsorbents

Adsorbent	**Isotherm**	**Max capacity (mg/g)**	**Kinetic Model**	**Referans**
Coffee husk	Sips	263	Pseudo second-order	[39]
Eleagnus stone	Langmuir	432.9	Pseudo second-order	[40]
Mango seed kerne	Langmuir	22.8	-	[41]
Pisum sativum L.	Freundlich	14.49	Pseudo second-order	[42]
Centaurea solstitialis	Temkin	122.5	Pseudo second-order	[26]

Walnut shell	Langmuir	90.8	Pseudo second-order	[43]
Sesame shell	Langmuir	1.58	Pseudo second-order	In this study

5. Energy Potential of Adsorbent

In the study on the usability of sesame stem as an energy source, its chemical properties were determined as 4.16% ash, 77.11% volatile matter, 14.54% fixed carbon, 4.19% moisture, and 4152 Cal/g [44]. In this study, the average value of 3618 Cal/g was found in the thermal value determination made without using sesame husk as an adsorbent. As it is known, efficient use of natural resources is important today. In this context, zero waste generation is extremely important in terms of sustainable environmental policy. It is aimed to use the waste adsorbent generated at the end of the study as a fuel in terms of sustainable environment. The thermal energy value of this adsorbent is measured. In the process performed on the calorimeter device, the thermal energy value of the adsorbent is selected as: 4122 Cal/g.

This result; shows that waste adsorbent can be used as fuel.

4. CONCLUSION

As a result of the study, it was found that the adsorption capacity of sesame peel used as adsorbent was q_{max}=1.58 mg/g, the adsorption fit the Langmuir isotherm model and the adsorption kinetics fit the pseudo second order kinetic model. The malachite green dye in the solution was removed from the water in 93.45% yield. According to the data obtained as a result of the study, it has been determined that SS can be used as an adsorbent with very high removal efficiency in the removal of MG dyestuff in aqueous solution. The disposal of the adsorbent with sustainable environmental mentality should also be taken into consideration and the calorific value of the adsorbent used in this context was examined and the calorific value of 4122 Cal/g revealed that it can be used in incineration plants.

REFERENCES

[1] S. Tang and M. A. A. Zaini, "Microporous activated carbon prepared from yarn processing sludge via composite chemical activation for excellent adsorptive removal of malachite green," *Surfaces and Interfaces,* vol. 22, no. 100832, pp. -, 2021.

[2] A. S. Sartepe, A. M. Mandhare, V. V. Jadhav, P. R. Raut, M. A. Anuse and S. S. Kolekar, "Removal of Malachite green dye from aqueous solution with adsorption technique using Limonia acidissima (Wood apple) shell as low cost adsorbent," vol. 10, pp. 3229-3238, 2017.

[3] P. Vandevivere, R. Bianchi and W. Verstraete, "Treatment and reuse of wastewater from the textile wet-processing industry: review of emerging technologies.," *J. Chem. Technol. Biotechnol. 72, 289-302.;,* Vols. -, no. 72, pp. 289-302, 1998.

[4] E. H. Robinson, M. H. Li and B. B. Manning, "Evaluation of corn gluten feed as a dietary ingredient for pond-raised channel catfish Ictalurus punctatus.," *J. World Aquacult. Soc.,* vol. 32, no. 1, pp. 68-71, 2001.

[5] H. Demiral, I. Demiral, F. Tümsek and B. Karabacakoğlu, "Pore structure of activated carbon prepared from hazelnut bagasse by chemical activation," *Surface and Interface Analysis,* Vols. -, no. 40, pp. 616-619, 2008/03/01.

[6] B. S. Karatas, E. Akkuzu, H. B. Unal, S. Aşik and M. Avci, "Using satellite remote sensing to assess irrigation performance in Water User Associations in the Lower Gediz Basin, Turkey," *Agricultural Water Management,* vol. 96, no. 6, pp. 982-990, 2009.

[7] F. P. Van der Zee and S. Villaverde, "Combined Anaerobic-Aerobic Treatment of Azo Dyes - A Short Review of Bioreactor Studies," *Water Research,* Vols. -, no. 39, pp. 1425-1440, 2005.

[8] M. Karatas, S. Dursun and M. Argun, "The decolourisation of azo dye Reactive Black 5 in a sequential anaerobic–aerobic system," *Ekoloji,* vol. 19, no. 74, p. 15–23, 2010.

[9] G. Kaykıoğlu, "Kolemanit ve Üleksit Atığı ile Sulu Çözeltilerden Metilen Mavisi Giderimi: Kinetik ve İzoterm Değerlendirmesi," *CBÜ Fen Bil. Dergi,* pp. 499-509, 2016.

[10] J. Nath, S. Bag, D. Bera and L. Ray, "Biotreatment of malachite green from aqueous solution and simulated textile effluent by growing cells (batch mode) and activated sludge system," *Groundwater for Sustainable Development,* vol. 8, pp. 172-178, 2019.

[11] "Mevzuat Bilgi Sistemi - Su Kirliliği Kontrolü Yönetmeliği," [Online]. Available: https://mevzuat.gov.tr/Metin.Aspx?MevzuatKod=7.5.7221&MevzuatIliski=0. [Accessed

16 11 2019].

[12] S. Dawood and T. Sen, "Review on dye removal from its aqueous solution into alternative cost effective and non-conventional adsorbents," *J. Chem. Process Eng.*, pp. 1-11, 2014.

[13] M. Gao, Z. Wang, C. Yang, J. Ning, Z. Zhou and G. Li, "Novel magnetic graphene oxide decorated with persimmon tannins for efficient adsorption of malachite green from aqueous solutions," *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, vol. 566, pp. 48-57, 2019.

[14] "Electrospun reduced graphene oxide/TiO2/poly(acrylonitrile-co-maleic acid) composite nanofibers for efficient adsorption and photocatalytic removal of malachite green and leucomalachite green," [Online]. Available: https://www.sciencedirect.com/science/article/pii/S0045653519320016#sec1.

[15] F. Guo, X. Jiang, X. Li, X. Jia, S. Liang and L. Qian, "Synthesis of MgO/Fe3O4 nanoparticles embedded activated carbon from biomass for high-efficient adsorption of malachite green," *Materials Chemistry and Physics*, vol. 240, 2020.

[16] W. Qu, T. Yuan, G. Yin, S. Xu, Q. Zhang and H. Su, "Effect of properties of activated carbon on malachite green adsorption," *Fuel*, vol. 249, pp. 45-53, 2019.

[17] S. C. Wei, S. Fan, C. W. Lien, B. Unnikrishnan, Y. S. Wang, H. W. Chu, C. C. Huang, P. H. Hsu and H. T. Chang, "Graphene oxide membrane as an efficient extraction and ionization substrate for spray-mass spectrometric analysis of malachite green and its metabolite in fish samples," *Analytica Chimica Acta*, vol. 1003, pp. 42-48, 2018.

[18] C. Akmil-Başar, Y. Önal, T. Kılıçer and D. Eren, "Adsorptions of high concentration malachite green by two activated carbons having different porous structures," *Journal of Hazardous Materials*, vol. 127, no. 1-3, pp. 73-80, 2005.

[19] E. Kuşvuran, O. Gülnaz, A. Samil and Ö. Yıldırım, "Decolorization of malachite green, decolorization kinetics and stoichiometry of ozone-malachite green and removal of antibacterial activity with ozonation processes," *Journal of Hazardous Materials*, vol. 1, no. 186, pp. 133-143, 2015.

[20] X.-J. Zhou, W.-Q. Guo, S.-S. Yang, H.-S. Zheng and N.-Q. Ren, "Ultrasonic-assisted ozone oxidation process of triphenylmethane dye degradation: Evidence for the promotion effects of ultrasonic on malachite green decolorization and degradation mechanism," *Bioresource Technology*, vol. 128, pp. 827-830, 2013.

[21] L. Man, P. Kumar, T. Teng and K. Wasewar, "Design of Experiments for Malachite Green Dye Removal from Wastewater Using Thermolysis-Coagulation-Flocculation," *Journal of Environmental Protection*, vol. 6, no. 12, pp. 260-271, 2012.

[22] L. Yong, G. Y. J. Zhanqi, H. Xiaobin, C. Sun, Y. L. W. Shaogui, W. Qingeng and F. Die, "Photodegradation of malachite green under simulated and natural irradiation: Kinetics, products, and pathways," *Journal of Hazardous Materials,* no. 285, pp. 127-136, 2015.

[23] "Brewers' spent grain in adsorption of aqueous Congo Red and malachite Green dyes: Batch and continuous flow systems," [Online]. Available: https://www.sciencedirect.com/science/article/pii/S0304389419308507#bib0005, .

[24] A. Eltaweil, H. Ali Mohammed, E. Abd El-Monaem and G. El-Subruiti, "Mesoporous magnetic biochar composite for enhanced adsorption of malachite green dye: Characterization, adsorption kinetics, thermodynamics and isotherms," *Advanced Powder Technology,* 2020.

[25] R. Bagheri, M. Ghaedi, A. Asfaram, E. A. Dil and H. Javadian, "RSM-CCD design of malachite green adsorption onto activated carbon with multimodal pore size distribution prepared from Amygdalus scoparia: Kinetic and isotherm studies," *Polyhedron,* vol. 171, pp. 464-472, 2019.

[26] M. Saleh, M. Yalvaç, H. Arslan and M. Gün, "Malachite Green Dye Removal from Aqueous Solutions Using Invader Centauera Solstitialis Plant and Optimization by Responce Surface Method: Kinetic, Isotherm, and Thermodynamic Study," *European Journal of Science and Technology,* no. 17, pp. 755-768, 2019.

[27] M. Saleh, M. Yalvaç and H. Arslan, "Optimization of Remazol Brilliant Blue R Adsorption onto Xanthium Italicum using the Response Surface Method," *Karbala International Journal of Modern Science,* vol. 5, no. 1, 2019.

[28] M. Bennet, "Sesame seed," THE NEW RURAL INDUSTRIES, [Online]. Available: https://www.agmrc.org/media/cms/sesame_38F4324EE52CB.pdf. [Accessed 15 03 2020].

[29] S. Kaya and T. Kahyaoğlu, "Influence of dehulling and roasting process on the thermodynamics of moisture adsorption in sesame seed," *Journal of Food Engineering,* vol. 76, no. 2, pp. 139-147, 2006.

[30] R. Ram, D. Catlin, J. Romero and C. Cowley, "Sesame: New Approaches for Crop Improvement," [Online]. Available: https://hort.purdue.edu/newcrop/proceedings1990/V1-225.html. [Accessed 15 03 2020].

[31] Türkiye İstatistik Kurumu, "Bitkisel Üretim İstatistikleri, 2020," Türkiye İstatistik Kurumu, Sayı: 33737, Ankara, 2020.

[32] E. Öztürk and G. Ova, "Yağlı Tohum Kabuklarının Biyoaktif Bileşen Potansiyeli ve Gıdalarda Kullanımı," *Akademik Gıda Dergisi, Ege Üniversitesi,* vol. 15, no. 3, pp. 315-321, 2017.

[33] A. Seçer, "Türkiye'de Susam Üretim ve Dış Ticaretinde Gelişmeler," *Çukurova Tarım Gıda Bilimleri Dergisi,* vol. 31, pp. 27-36, 2016.

[34] A. S. Eltaweil, H. A. Mohammed, E. M. A. El-Monaem and G. M. El-Subruiti, "Mesoporous magnetic biochar composite for enhanced adsorption of malachite green dye: Characterization, adsorption kinetics, thermodynamics and isotherms," *Advenced Powder Technology,* 2020.

[35] A. Yıldırım and Y. Bulut, "Adsorption behaviors of malachite green by using crosslinked chitosan/ polyacrylic acid/ bentonite composites with different ratios," *Environmental Technology & Innovation,* vol. 17, 2020.

[36] S. Savcı, M. Yalvaç and Z. Karaman, "Adsorption of Remazol Brilliant Blue R from Aquatic Solution Using Natural Adsorbent (Pistachio hull)," *Ecological Life Sciences,* vol. 13, no. 2, pp. 103-113, 2018.

[37] T. K. Arumugam, P. Krishnanmoorthy, N. R. Rajagopalan, S. Nanthini and D. Vasudevan, "Removal of malachite green from aqueous solutions using a modified chitosan composite," *International Journal of Biological Macromolecules,* vol. 128, pp. 655-664, 2019.

[38] T. Büyüksırıt and H. Kuleaşan, "Fourier Dönüşümlü Kızılötesi (FTIR) Spektroskopisi ve Gıda Analizlerinde Kullanımı," *Hitit Üniversitesi Gıda Dergisi (GIDA),,* vol. 39, no. 4, pp. 235-241, 2014.

[39] T. Murthy, B. Gowrishankar, M. Parabha, M. Kruthi and R. Krishna, "Studies on Batch Adsorptive Removal of Malachite Green from Synthetic Wastewater Using Acid Treated Coffee Husk: Equilibrium, Kinetics and Thermodynamic Studies," *Microchemical Journal,* vol. 146, pp. 192-201, 2019.

[40] G. Gökara, "İğde Çekirdeğinden Elde Edilen Aktif Karbon Üzerinde Tekstil Boyar Maddelerinin Adsorpsiyonu," Trakya Üniversitesi, Fen Bilimleri Enstitüsü, Fizikokimya Anabilim Dalı, Edirne, 2013.

[41] D. Singh, V. Sowmya, S. Abinandan and S. Shanthakumar, "Removal of Malachite Green Dye by Mangifera İndica Seed Kernel Powder," *Journal Institution of Engineers,* vol. 99, no. 1, pp. 103-111, 2018.

[42] A. Tabrez, R. Rumana, A. Imran, E. A.K. and A. Amer, "Removal of Malachite Green From Aqueous Solution Using Waste Pea Shells as Low-Cost Adsorbent – Adsorption İsotherms and Dynamics," *Toxicological & Environmental Chemistry,* vol. 96, no. 4, pp. 569-578, 2014.

[43] M. K. Dahri, M. R. Kooh and L. B. Lim, " Water remediation using low cost adsorbent walnut shell for removal of malachite green: Equilibrium, kinetics, thermodynamic and

regeneration studies," *Journal of Environmental Chemical Engineering,* vol. 2, pp. 1434-1444, 2014.

[44] Gebresas, Alula; Asmelash, Haftom; Berhe, Hadush; Tesfay, Tsegay;, "Briquetting of Charcoal from Sesame Stalk," *Journal of Energy,* vol. 2015, no. 757284, p. 6, 2015.

[45] S. H. Tang and M. A. Ahmad Zaini, "Development of activated carbon pellets using a facile low-cost binder for effective malachite green dye removal," *Journal of Cleaner Production,* vol. 253, 2020.

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