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The Effective and Eco-friendly Tea Fungus for the Biosorption of Dye Pollutant from Aqueous Solutions

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Abstract

The present study has reported the production of the environmentally friendly Kombucha tea fungus (TF), as well as its investigation as a potential biosorbent to remove cationic methyl violet (MV) dye from the aqueous solutions. TF was characterized by using FT-IR, thermal analysis and zeta potential measurements. The biosorption of MV was significantly dependent on the pH of the solutions and the highest removal for MV was observed at pH=9.0 which was also proved with zeta potential measurement. Biosorption equilibrium was established in 10 min and a definition of the overall rate-controlling step in the biosorption of MV onto TF was carried out. The biosorption data were in good agreement with the pseudo-second-order kinetic and Langmuir isotherm models. The maximum biosorption capacity of TF for MV was determined as 1180.09 mg g^{-1}. In addition, an effective biosorption performance was observed even in the presence of high foreign ion concentrations. The results indicated that TF could be utilized as a highly efficient biosorbent for MV biosorption from aqueous solutions.

Keywords: Tea fungus; Biosorption; Methyl violet; Isotherms.

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Sulu Çözeltilerdeki Boyarmadde Kirliliğinin Biyosorpsiyonu için Etkili ve Çevre Dostu Çay Mantarı

Öz

Bu çalışma, çevre dostu Kombucha çay mantarının (TF) üretimini ve sulu çözeltilerden katyonik metil viyole (MV) boyarmaddesinin uzaklaştırılmasında biyosorban olarak potansiyelinin araştırılmasını sunmaktadır. TF, FT-IR, termal analiz ve zeta potansiyel ölçümleri kullanılarak karakterize edilmiştir. MV'nin biyosorpsiyonu, çözeltilerin pH'sına önemli ölçüde bağlıdır ve en yüksek MV giderimi, zeta potansiyeli ölçümü ile de kanıtlanan pH=9.0'da gözlenmiştir. Biyosorpsiyon 10 dakika içerisinde dengeye ulaşmış ve MV'nin TF üzerine biyosorpsiyonunda hız sınırlayıcı adım belirlenmiştir. Biyosorpsiyon verileri, yalancı ikinci dereceden kinetik ve Langmuir izoterm modelleri ile iyi bir uyum göstermiştir. TF'nin MV için maksimum tek tabakalı biyosorpsiyon kapasitesi 1180.09 mg g^{-1} olarak belirlenmiştir. Ayrıca, yüksek yabancı iyon derişimlerinin varlığında dahi etkili bir biyosorpsiyon performansı gözlenmiştir. Sonuçlar, TF'nin sulu çözeltilerden MV biyosorpsiyonu için yüksek verimli bir biyosorban olarak kullanılabileceğini göstermiştir.

Anahtar Kelimeler: Çay mantarı; Biyosorpsiyon; Metil viyole; İzotermler.

1. Introduction

Kombucha is a traditional probiotic drink that is prepared from the fermentation of sugar added black or green tea with lactic acid bacteria, acetic bacteria, and yeasts [1-3]. After fermentation, a symbiotic culture namely, *Medusomyces gisevii* (tea fungus) layer forms that floats on the tea in 7-15 days [4]. Under the culture, there is Kombucha tea with a pH in the range of 2.5-3.0. Kombucha tea is originated from China (Manchuria) and after gained rapid popularity and spread all over the world, mainly to Russia and Germany [5]. Kombucha tea contains several components with known benefits for human health such as water-soluble vitamins, minerals, amines, purines, flavonoids, catechins, tannins, essential oils and hydrolytic enzymes [6-8]. In particular, it has anti-diabetic, anti-carcinogenic and anti-inflammatory effects. Moreover, it plays a healing role in the treatment of gastric ulcers, lowering high cholesterol, improvement of the liver and the immune system [9]. As the demand for Kombucha increases, to gain more benefits from obtained culture, the application potential in various industrial areas such as textiles and cosmetics has been investigated [10-12]. In addition, the use of this fungus to eliminate the environmental damage of pollutants in industrial wastewater has attracted attention as a popular biotechnological research subject. Among the pollutants, dyes are substances that have a high potential for use in most various industrial areas such as food, leather, textile, paper and plastics [13,14]. Methyl violet (MV) which is a cationic dye with high color intensity is extensively used as a purple dye for textile products such as silk, wool, nylon, cotton and in printing ink. In addition, MV has been widely utilized as a pH indicator [15]. It is employed as an ingredient in Gram's stain for classifying bacteria and also a bacteriostatic agent in the medical field. MV has a toxic organic structure which causes diverse health problems for humans such as cancer, skin irritation, respiratory and kidney failure and permanent blindness [16, 17]. Because of all its hazard mentioned above, MV needs to be effectively removed from the aqueous solutions before being discharged into the environment. There are diverse methods such as biological, physical and chemical that are utilized to eliminate pollutants from the aqueous solutions [18, 19]. However, their uses have many restraints such as application difficulty, process cost, low yield and occurrence of huge amount of sludge. Biosorption is a popular pollution treatment method that is ecofriendly, economic, easy to application, non-toxic and high efficiency [20, 21]. Biological origin materials such as bacteria [22], fungus [23], alg [24] and agricultural wastes [25] are used as biosorbents in the biosorption.

In this context, tea fungus (TF) was used as a biosorbent for the biosorption of methyl violet (MV) from the aqueous solutions. The characterization of the TF was carried out with Infrared Spectroscopy (IR), Thermogravimetric (TG) analysis and zeta potential measurements. The effects of pH, TF amount, temperature, contact time, initial MV concentration and ionic strength on the biosorption were investigated to determine the best biosorption conditions. The biosorption kinetics and equilibrium isotherms data were modeled and the mechanism of the biosorption process was evaluated. The maximum biosorption capacity of TF towards MV was compared with other studies in the literature.

2. Materials and Methods

2.1. Chemicals and preparation of TF

MV was used as a dye pollutant in the biosorption process. Its maximum absorption wavelength (λ_{max}) and molecular weight were 584 nm and 393.9 g mol^{-1}, respectively. HCl and NaOH solutions in various concentrations were preferred to adjust the pH values. Deionized water was utilized for dissolving dye and preparing the target concentration of solutions. All the reagents provided for the biosorption were analytical grade and purchased from Merck. The Kombucha tea with fungus, sugar and black tea leaves were provided from the local market in Turkey. The production process of the fungus in biosorption included the following steps.; First, 3 L of water was put in a glass bottle and boiled, then 45 g L^{-1} sugar was added into this water. After the sugar was dissolved, 10 g of black tea leaves were added into the solution and brewed for 15 min. Subsequently, the solid-liquid phase was separated by filtration and the solution was kept in a glass bottle. Kombucha tea and fungus were added to the previous solution at room temperature and then left to the incubation process for about 15 days. At the end of this process, a disc-shaped, smooth new fungus (TF) was obtained on the Kombucha tea. Before being used in the biosorption process, this fungus was washed with plenty of deionized water to eliminate any possible impurities. It was put in an oven at 65ºC for drying. Then, it was ground and sieved through a 200 µm sieve to obtain a homogeneous grain-sized biosorbent (Fig. 1). Finally, it was stored in a capped bottle for utilization in all experiments.

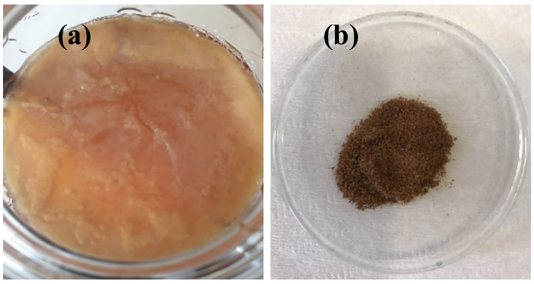

Figure 1: Kombucha tea fungus before (a) and after (b) drying, grinding and sieving

2.2. Biosorption studies

The various parameters (pH, contact time, biosorbent amount, temperature, initial pollutant concentration and ionic strength) were investigated for the biosorption of MV. The pH experiments were carried out by 20 mL of a 100 mg L^{-1} MV solution with 0.625 g L^{-1} of TF and the pH value was attentively adjusted between 2.0 and 10.0 by adding a small volume of NaOH or HCl solutions using a Hanna pH meter. MV solutions were stirred in a 25 mL glass bottle for 60 min. To determine the optimum biosorbent amount, TF was weighed from 0.250 to 1.25 g L^{-1} and added into 100 mg L^{-1} MV at pH 9.0. The temperature was changed between 15 and 40ºC at an initial MV concentration of 100 mg L^{-1}. In order to execute kinetic analysis, contact time for MV biosorption onto TF was applied in the range of 1–90 min. The effect of initial MV concentration on the biosorption capacity of TF was examined by changing the concentrations from 50 to 800 mg L^{-1} at the confirmed conditions. To investigate the ionic strength effect on the biosorption of MV, the biosorption experiments were occurred with 100 mg L^{-1} of MV solutions containing NaCl in the concentration range of 0.02−0.1 mol L^{-1} under the optimum biosorption conditions. After each application of biosorption experiment, the TF was separated from the medium via filtration. All experiments were repeated in triplicate. The obtained samples were measured by using a UV-vis spectroscopy (Shimadzu UV-2600 spectrophotometer). The obtained data were evaluated by biosorption amount and biosorption (%) of MV using the equations which were given as below

$$q_e = \frac{V \times (C_0 - C_e)}{m} \tag{1}$$

$$\text{Biosorption (\%)} = \frac{C_0 - C_e}{C_0} \tag{2}$$

In this equation, q_e: The amount of MV biosorbed on TF at equilibrium (mg g^{-1}), m: The amount of TF (g), C_0: The initial concentration (mg L^{-1}) of MV, C_e: equilibrium concentration (mg L^{-1}) of MV, V: The volume of MV solution (L).

2.3. Characterizations

FT-IR analyses were (Bruker Tensor 27 FT–IR spectrometer) carried out in the 400–4000 cm^{-1} range to identify the surface functional groups of TF before and after MV biosorption. Thermogravimetric analysis (TGA) of TF in the temperature range of 30–700ºC was done by using Perkin Elmer Diamond TG/DTA instrument with a rate of 10ºC min^{-1} under the air atmosphere. The pH-dependent surface charges of TF were specified using a zeta potential measurement (ZEN 3600 Model Zetasizer Nano-ZS). For this measurement, 0.50 g L^{-1} of TF was added into 20 mL of deionized water and the pH was changed in the range of 2-10.

3. Results and Discussion

3.1. Characterization of FT

TF was characterized by FT-IR spectroscopy and thermal analysis. As mentioned in the introduction, TF has several components in its ingredient. In the FT-IR spectrum of TF, the broad band appearing at 3381 cm^{-1} can be owing to ν(O-H) stretching vibrations of phenolic alcohols. Aliphatic ν(C-H) stretching vibrations appear at 2937 cm^{-1} and 2898 cm^{-1}. The picks observed at 1727 cm^{-1} and 1638 cm^{-1} are assigned to the carbonyl ν(C=O) and imine ν(C=N) stretching vibrations, respectively. Aromatic ν(C=C) stretching vibration is observed at 1424 cm^{-1}. After MV biosorption, the pick observed at 3381 cm^{-1} shifts to high wavenumber and also the intensity of aliphatic ν(C-H) stretching vibration decreases. They can be due to the fact that phenolic –OH, imine and aliphatic –CH- groups are covered by dye molecules. The carbonyl pick at 1727 cm^{-1} disappears and some peaks shift (Fig. 2). They show the biosorption of MV onto TF. TG/DTG curves are acquired to determine the decomposition stage of TF. As illustrated in Fig. 3, the first stage in the temperature range 30-100ºC is the elimination of water molecules. After this step, the

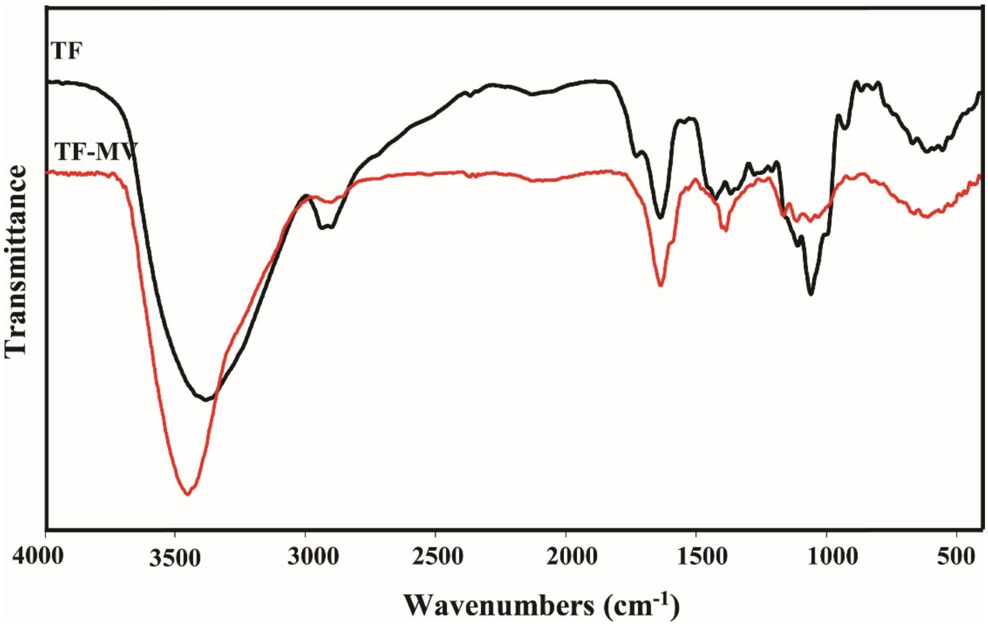

Figure 2: FT-IR spectra of TF and TF-MV

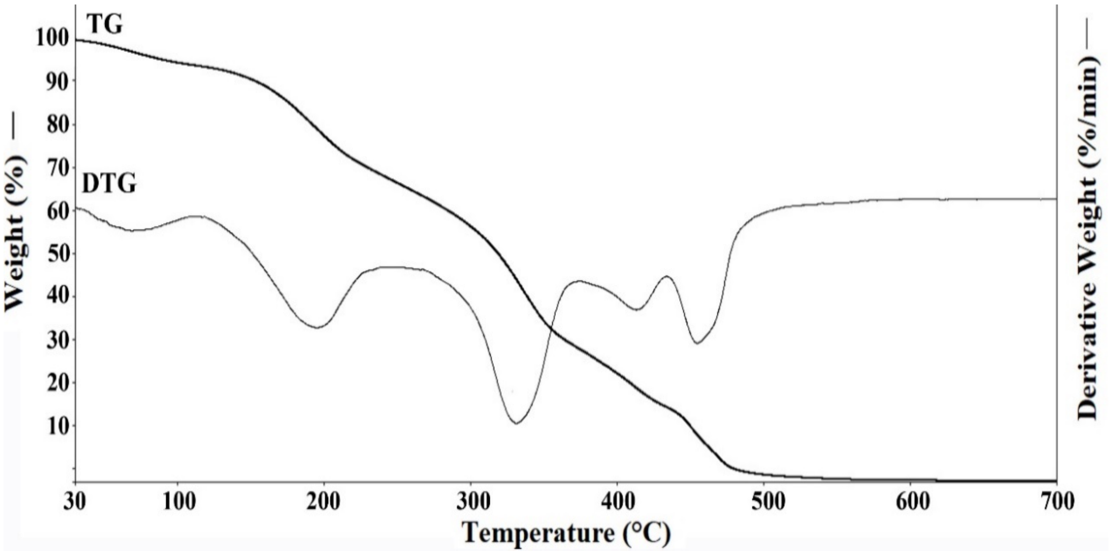

Figure 3: TG/DTG curves of TF

3.2. Biosorption parameters

3.2.1. Effect of pH

Biosorption amount is generally affected by the pH of the dye solution because of affecting the functional groups on the biosorbent and the ionization degree of the dye molecules. Thus, it is a major experimental parameter that should be considered. Fig. 4 displays the results for the biosorption of MV onto TF when the pH is changed at a range of 2.0–10.0 of the MV solutions at room temperature. The biosorption capacity of TF for MV removal increases with the increasing pH of the biosorption medium and reaches an equilibrium value of 154.46 mg g^{-1} at pH 9.0 (Fig. 4). After this pH value, no significant change appears in the MV biosorption ability of TF. Therefore, pH 9.0 is selected as the pH value of the biosorption media for further experiments. As the acidity of the biosorption medium increases, the positive charge density increases due to the protonation of the functional groups on the biosorbent surface. Thus, the interaction of cationic dye molecules with the biosorbent surface is restricted, resulting in lower biosorption. The negative charge density related to the concentration of OH^- ions rises on the biosorbent surface when the pH increases. Thus, the electrostatic interactions between cationic dye MV and TF are enhanced. The surface charge density values of TF in deionized water at various pHs which support the obtained data from the pH parameter are presented in Fig. 4. It is showed that the zeta potential of TF decreases considerably from −1.60 mV at pH 2.0 to −30.40 mV at pH 10.0. TF has no points of zero charge and the biosorbent surface has the highest negative charge density (−35.40 mV) at pH=9.0 where the maximum amount of the biosorption is achieved.

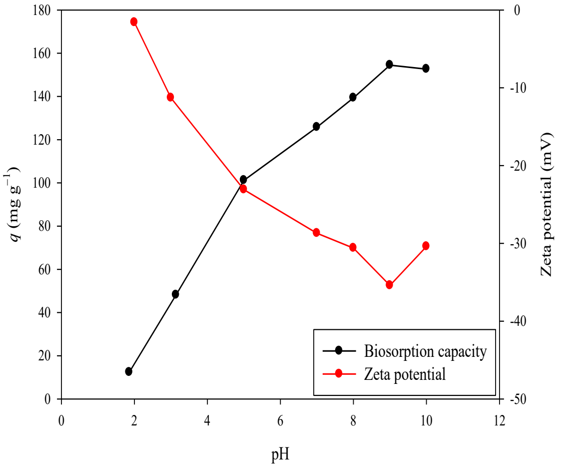

Figure 4: Zeta potential of TF in deionized water and pH effect on the MV biosorption

3.2.2. Effect of TF amount and temperature

The biosorbent amount is another important parameter that highly affects the biosorption process yield and cost. It has been indicated that the pollutants are removed at high efficiency with the increase in the biosorbent amount which gives rise to the increase of active sites on the biosorbent surface [26]. Hence, the effect of TF amount on the MV biosorption was examined in the range of 0.25–1.00 g L^{-1} with pH of 9.0 (Fig 5a). As seen in Fig 5a, the biosorption yield increases from 84.75% to 96.83% as the amount of TF increases from 0.250 to 0.500 g L^{-1}. It is noted that after 0.750 g L^{-1} amount of TF, the biosorption yield decreases to 86.66%. This decrease can be explained with the fact/inference that the biosorbent can aggregate at higher amounts and its active binding sites can be lower for interaction with dye molecules. With a very economic amount of 0.500 g L^{-1}, the biosorption reaches its highest efficiency. For this reason, the optimum amount of TF to be used in further biosorption experiments was determined as 0.500 g L^{-1}.

To investigate whether the biosorption was temperature-dependent, the biosorption experiments were realized at the temperature range of 15–40ºC at the initial MV concentration of 100 mg L^{-1}, TF amount of 0.500 g L^{-1} and the contact time of 60 min. As depicted in Fig. 5b, the biosorption amounts of MV change from 190.20 to 191.10 mg g^{-1} with the increase of the temperature from 15 to 40ºC. The results show an insignificant effect of temperature in the biosorption ability of TF for MV removal. Similar results for the effect of temperature are observed in the literature [27-28]. As a result, this system is very advantageous as it has the potential for the elimination of MV from the solution with very high efficiency at any temperature.

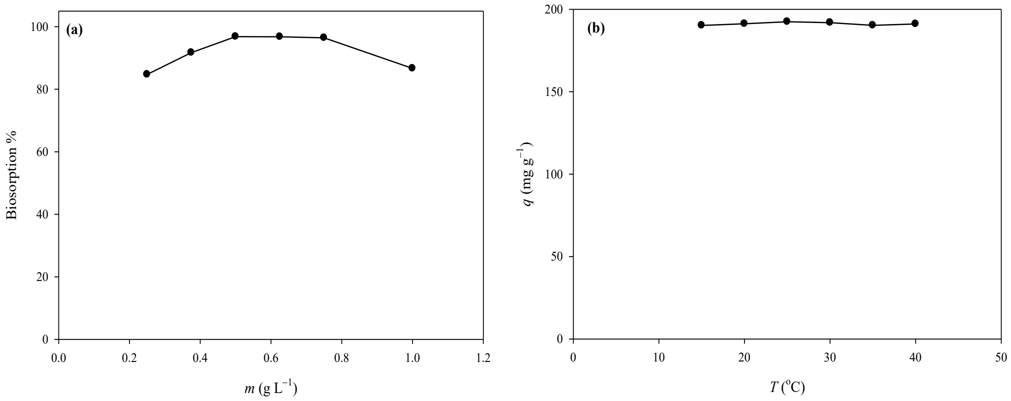

Figure 5: The effect of TF amount (a) and temperature (b) on the MV biosorption

3.2.3. Effect of contact time and biosorption kinetics

To provide information about the biosorption behavior of MV onto TF as a function of contact time, the biosorption experiments were performed in the contact time range of 1–90 min at 20ºC. The effect of contact time on biosorption is demonstrated in Fig 6a. MV is rapidly removed from the aqueous solution in 10 min and after this point, the biosorption equilibrium is established. This rapid biosorption is due to the binding of the MV molecules to a great number of vacant active biosorption sites on TF. The biosorption amount of MV rises from 180.52 to 188.86 mg g^{-1} in the time range of 1-10 min. The biosorption reaching equilibrium quite fast is an important advantage that supports the practicality of the method. A similar study was also reported for biosorption equilibrium time as 10 min [29]. The biosorption kinetic applications are commonly utilized to reveal the control mechanism of the biosorption process.

In this study, to explore the MV biosorption nature, the obtained experimental data from contact time were applied to Lagergren-first-order [30], pseudo-second-order [31] and intra-particular diffusion kinetic models [32]. The kinetic parameters obtained from fitting the experimental data to these three kinetic model equations are presented in Table 1. When the highest R^2 values are compared, it is easy to see that the R^2 of the pseudo-second-order model for TF (R^2= 0.999) is significantly higher than Lagergren-first-order (R^2= 0.597) and intra-particular diffusion kinetic models (R^2= 0.787). The plot of the pseudo-second-order kinetic model for the biosorption of MV onto TF is exhibited in Fig. 6b. The obtained data for the biosorption of MV onto TF are better fitted to the pseudo-second-order kinetic model. This finding indicates that the rate-limiting step in the biosorption process can be chemical biosorption which includes electron exchange or electron sharing between MV and TF.

Table 1: Kinetic parameters for the biosorption of MV

Model	**Parameters**	**Values**
Lagergren-first-order model	k_1	1.97×10^{-2}
$\log(q_e - q_t) = \log(q_e) - \frac{k_1}{2.303}t$	q_e	7.96
	R^2	0.597
Pseudo-second-order model	k_2	1.31×10^{-2}
$\frac{t}{q_t} = \frac{1}{k_2 {q_e}^2} + \left(\frac{1}{q_e}\right)t$	q_e	194.18
	R^2	0.999
Intraparticle diffusion model	k_p	3.11
$q_t = K_p t^{1/2} + C$	C	178.94
	R^2	0.787

(where, q_t (mg g^{-1}): The biosorption capacity of TF at t time, k_1 (min^{-1}), k_2 (g mg^{-1} min^{-1}), k_p (mg g^{-1} $min^{-1/2}$): The rate constants, C (mg g^{-1}): The intercept.)

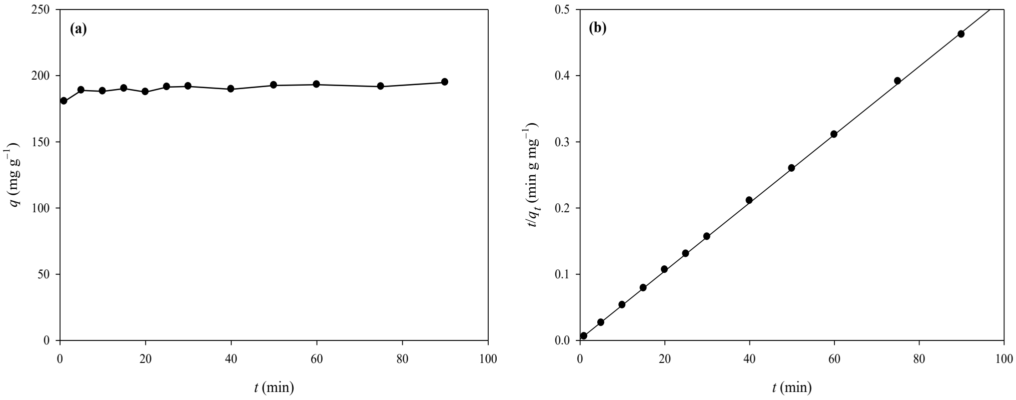

Figure 6: The effect of contact time (a) and the plot of the pseudo-second-order kinetic model for the MV biosorption (b)

3.2.4. Effect of initial MV concentration and biosorption isotherms

Various isotherm models used to interpret experimental biosorption data at equilibrium give significant information about the biosorption mechanism and maximum biosorption capacity of the biosorbent. In this study, the obtained data in equilibrium were applied to Langmuir [33] and Freundlich [34] isotherm models. According to the general isotherm plot (Fig. 7a), the obtained data prove that the biosorption capacity of TF increases from 87.20 to 1149.0 mg g^{-1} with increasing the initial concentration of MV from 50 to 800 mg L^{-1} in the solutions. This can be due to the occupation of the biosorption sites on TF as the concentration of the MV increases. The biosorption process reaches equilibrium when the MV concentration is 700 mg L^{-1} and after this concentration value, the biosorption amount of MV is constant.

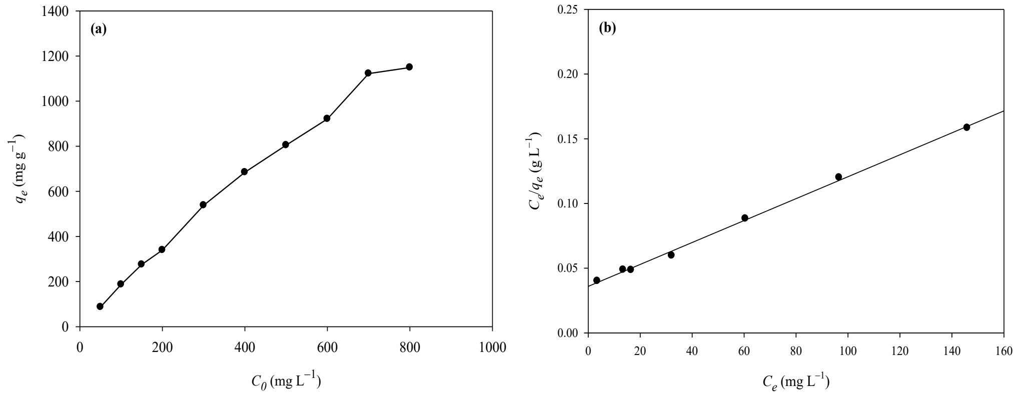

Figure 7: General isotherm plot (a) and Langmuir isotherm model for the MV biosorption (b)

Table 2 shows the Langmuir and Freundlich isotherm parameter values for MV biosorption onto TF. From the results, the data are well fitted to the linearized Langmuir isotherm model than the Freundlich isotherm model because of the highest R^2 value. The values of q_{max} and K_L are identified from the slope and intercept of the linear Langmuir plot of C_e/q_e versus C_0 (Fig 7b). The maximum biosorption capacity of TF is determined as 1180.09 mg g^{-1} for MV cationic dye from this model. The R_L value of 0.175 demonstrates the favorable interaction between the TF surface and MV molecules. From the results, it can be assumed that the maximum biosorption amount corresponds to saturated monolayered MV molecules on the TF surface with constant energy. The maximum biosorption capacity of TF for MV is quite high or comparable when compared to several biosorbents in the literature (Table 3). TF is considered environmentally friendly, as it not only has an excellent biosorption capacity but also is not treated with any chemicals.

Table 2: Isotherm parameters for the biosorption of MV

Isotherm model	Equations	Parameters	R^2
Langmuir	$\frac{C_e}{q_e} = \frac{1}{q_{max}K_L} + \frac{C_e}{q_{max}}$ $R_L = \frac{1}{1 + K_L C_0}$	q_{max}=1180.09, K_L=2.36×10^{-2} R_L=0.175	0.998
Freundlich	$\ln q_e = \ln K_F + \frac{1}{n}\ln C_e$	n=1.608, K_F=50.60	0.956

(where, q_{max}: The maximum MV biosorption capacity of TF (mg g^{-1}), K_L and K_F: The Langmuir (L mg^{-1}) and Freundlich (L g^{-1}) constants, respectively, R_L: The separation factor constant and n: Heterogeneity factor.)

Table 3: Comparison of biosorption capacity of TF for MV with other sorbents reported previously

Biosorbent	q_m (mg g^{-1})	Reference
Calcined lotus leaf	26.315	[35]
PFPCS	3756.33	[36]
Dithiocarbamate-grafted star-like polymer	1239	[37]
Graphene oxide hydrogel composite	1052.63	[38]
Carya illinoensis powder	642.0	[39]
Anethum graveolens	833.33	[40]
Pu-erh tea powder	277.78	[41]
Breadfruit core	307	[42]
Phragmites australis activated carbon	351.33	[43]
TF	1180.09	Present study

3.2.5. Ionic strength effect

The ionic strength effect on the MV biosorption onto TF was examined under optimum conditions (pH: 9, TF amount: 0.500 g L^{-1}, t: 10 min, initial MV concentration: 100 mg L^{-1}, T: 20ºC). For this purpose, NaCl solutions were added to MV solutions in such a way that the final salt concentrations in the solutions were between 0.02 mol L^{-1} and 0.1 mol L^{-1}. As seen in Figure 8, the biosorption capacity of TF results in a reduction of approximately 12% when the salt concentration is 0.02 mol L^{-1} and the biosorption efficiency of MV gradually decreases with increasing NaCl concentration. The biosorption yield of MV is evaluated as 72.20% when the concentration of NaCl reaches 0.08 mol L^{-1} and after this point, the increase in the salt concentration does not influence the biosorption. This can be expressed by a competition between cationic MV molecules and Na^+ ions, which decreases the electrostatic charge on the surface or raises ionic strength [42-43].

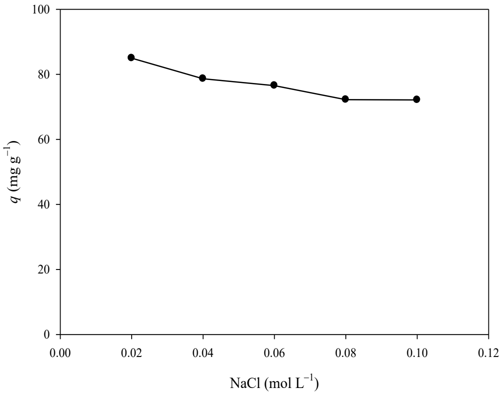

Figure 8: The ionic strength effect on the MV biosorption

4. Conclusion

Kombucha tea fungus (TF) was simply prepared with sugar, black tea leaves and tea containing Kombucha culture. It was used as a biosorbent for methyl violet (MV) biosorption from aqueous solutions. TF was identified with various characterization techniques such as FT-IR, TG analysis and zeta potential measurements. The results demonstrated that pH was an important parameter on the biosorption of MV. The maximum biosorption capacity for TF for the biosorption of MV was achieved at pH=9.0 and the biosorption amount of MV was evaluated as 188.86 mg g^{-1} in a short 10 min contact time with 0.500 g L^{-1} of TF. The observed extremely short equilibrium time is a significant advantage in saving time in the biosorption process. The changes in temperature from 15 to40ºC did not lead to any changes in the MV biosorption. The biosorption process was well defined with the Langmuir isotherm model and the calculated maximum biosorption capacity value was 1180.09 mg g^{-1}. As a result, TF was superior to many biosorbents because of its high biosorption capacity for MV and also its being a green biosorbent that did not undergo any chemical treatment. All these findings have proven that TF was very effective in dye pollutant removal as well as in other areas of use in industry.

References

[1] Marsh, A.J., O'Sullivan, O., Hill, C., Ross, R.P., Cotter, P.D., *Sequence-based analysis of the bacterial and fungal compositions of multiple kombucha (tea fungus) samples,* Food Microbiology, 38, 171-178, 2014.

[2] Dutta, H., Paul, S.K., *Kombucha drink: production, quality, and safety aspects,* Production and Management of Beverages, 1, 259-288, 2019.

[3] Taheur, F.B., Mansour, C., Jeddou, K.B., Machreki, Y., Kouidhi, B., Abdulhakim, J.A., Chaieb, K., *Aflatoxin B1 degradation by microorganisms isolated from Kombucha culture,* Toxicon, 179, 76-83, 2020.

[4] Hesseltine, C.W., *A millennium of fungi, food, and fermentation,* Mycologia, 57, 149-197, 1965.

[5] Blanc, P.J., *Characterization of the tea fungus metabolites,* Biotechnology Letters, 18, 139-142, 1996.

[6] Pasha, C., Reddy, G., *Nutritional and medicinal improvement of black tea by yeast fermentation,* Food Chemistry, 89, 449-453, 2005.

[7] Malbaša, R.V., Lončar, E.S., Vitas, J.S., Čanadanović-Brunet, J.M., *Influence of starter cultures on the antioxidant activity of kombucha beverage,* Food Chemistry, 127, 1727-1731, 2011.

[8] Velićanski, A., Cvetković, D., Markov, S., *Characteristics of Kombucha fermentation on medicinal herbs from Lamiaceae family,* Romanian Biotechnological Letters, 18, 8034-8042, 2013.

[9] Villarreal-Soto, S.A., Beaufort, S., Bouajila, J., Souchard, J.P., Taillandier, P., *Understanding kombucha tea fermentation: a review,* Journal of Food Science, 83, 580-588, 2018.

[10] Amarasekara, A.S., Wang, D., Grady, T.L., *A comparison of kombucha SCOBY bacterial cellulose purification methods,* SN Applied Sciences, 2, 1-7, 2020.

[11] Dima, S.-O., Panaitescu, D.-M., Orban, C., Ghiurea, M., Doncea, S.-M., Fierascu, R.C., Nistor, C.L., Alexandrescu, E., Nicolae, C.-A., Trică, B., *Bacterial nanocellulose from side-streams of kombucha beverages production: Preparation and physical-chemical properties,* Polymers, 9, 374, 2017.

[12] Soares, M.G., de Lima, M., Schmidt, V.C.R., *Technological aspects of kombucha, its applications and the symbiotic culture (SCOBY), and extraction of compounds of interest: A literature review,* Trends in Food Science & Technology, 110, 539-550, 2021.

[13] Srivastava, S., Sinha, R., Roy, D., *Toxicological effects of malachite green,* Aquatic Toxicology, 66, 319-329, 2004.

[14] Al-Fawwaz, A.T., Abdullah, M., *Decolorization of methylene blue and malachite green by immobilized Desmodesmus sp. isolated from North Jordan,* International Journal of Environmental Science and Development, 7, 95, 2016.

[15] Bale, M., *Management of the umbilicus with crystal violet solution,* Canadian Medical Association Journal, 124, 372, 1981.

[16] Mittal, A., Gajbe, V., Mittal, J., *Removal and recovery of hazardous triphenylmethane dye, Methyl Violet through adsorption over granulated waste materials,* Journal of Hazardous Materials, 150, 364-375, 2008.

[17] Parshetti, G., Saratale, G., Telke, A., Govindwar, S., *Biodegradation of hazardous triphenylmethane dye methyl violet by Rhizobium radiobacter (MTCC 8161),* Journal of Basic Microbiology, 49, S36-S42, 2009.

[18] Rott, U., Minke, R., *Overview of wastewater treatment and recycling in the textile processing industry,* Water Science and Technology, 40, 137-144, 1999.

[19] El Haddad, M., Slimani, R., Mamouni, R., Laamari, M.R., Rafqah, S., Lazar, S., *Evaluation of potential capability of calcined bones on the biosorption removal efficiency of safranin as cationic dye from aqueous solutions,* Journal of the Taiwan Institute of Chemical Engineers, 44, 13-18, 2013.

[20] Wang, J., Chen, C., *Biosorbents for heavy metals removal and their future,* Biotechnology Advances, 27, 195-226, 2009.

[21] Mosier, A.P., Behnke, J., Jin, E.T., Cady, N.C., *Microbial biofilms for the removal of* Cu^{2+} *from CMP wastewater,* Journal of Environmental Management, 160, 67-72, 2015.

[22] Nguyen, T.A., Fu, C.-C., Juang, R.-S., *Biosorption and biodegradation of a sulfur dye in high-strength dyeing wastewater by Acidithiobacillus thiooxidans,* Journal of Environmental Management, 182, 265-271, 2016.

[23] Fan, H., Yang, J., Gao, T., Yuan, H., *Removal of a low-molecular basic dye (Azure Blue) from aqueous solutions by a native biomass of a newly isolated Cladosporium sp.: kinetics, equilibrium and biosorption simulation,* Journal of the Taiwan Institute of Chemical Engineers, 43, 386-392, 2012.

[24] Moghazy, R.M., Labena, A., Husien, S., *Eco-friendly complementary biosorption process of methylene blue using micro-sized dried biosorbents of two macro-algal species (Ulva fasciata and Sargassum dentifolium): Full factorial design, equilibrium, and kinetic studies,* International Journal of Biological Macromolecules, 134, 330-343, 2019.

[25] Ezeonuegbu, B.A., Machido, D.A., Whong, C.M., Japhet, W.S., Alexiou, A., Elazab, S.T., Qusty, N., Yaro, C.A., Batiha, G.E.-S., *Agricultural waste of sugarcane bagasse as efficient adsorbent for lead and nickel removal from untreated wastewater: Biosorption, equilibrium isotherms, kinetics and desorption studies,* Biotechnology Reports, 30, e00614, 2021.

[26] Sintakindi, A., Ankamwar, B., Fungal biosorption as an alternative for the treatment of dyes in waste waters: a review, Environmental Technology Reviews, 10, 26-43, 2021

[27] Alp Arici, T., *Highly reusable plant-based biosorbent for the selective methylene blue biosorption from dye mixture in aqueous media,* International Environmental Science and Technology, 1-12, 2021. https://doi.org/10.1007/s13762-021-03238-w

[28] Yang, Y., Wei, X., Sun, P., Wan, J., *Preparation, characterization and adsorption performance of a novel anionic starch microsphere,* Molecules, 15, 2872-2885, 2010.

[29] Deniz, F., Kepekci, R.A., *Dye biosorption onto pistachio by-product: A green environmental engineering approach,* Journal of Molecular Liquids, 219, 194-200, 2016.

[30] Lagergren, S., *Zur theorie der sogenannten adsorption geloster stoffe*, Kungliga Svenska Vetenskapsakademiens. Handlingar, 24, 1-39 1898.

[31] Ho, Y.-S., McKay, G., *Kinetic models for the sorption of dye from aqueous solution by wood,* Process Safety and Environmental Protection, 76, 183-191, 1998.

[32] Weber Jr, W.J., Morris, J.C., *Kinetics of adsorption on carbon from solution,* Journal of the Sanitary Engineering Division, 89, 31-59, 1963.

[33] Langmuir, I., *The adsorption of gases on plane surfaces of glass, mica and platinum,* Journal of the American Chemical society, 40, 1361-1403, 1918.

[34] Freundlich, H., *Über die adsorption in lösungen, Zeitschrift für physikalische Chemie,* 57, 385-470, 1907.

[35] Sharafzad, A., Tamjidi, S., Esmaeili, H., *Calcined lotus leaf as a low-cost and highly efficient biosorbent for removal of methyl violet dye from aqueous media,* International Journal of Environmental Analytical Chemistry, 1-24, 2020.

[36] Chen, K., Du, L., Gao, P., Zheng, J., Liu, Y., Lin, H., *Super and selective adsorption of cationic dyes onto carboxylate-modified passion fruit peel biosorbent,* Frontiers in Chemistry, 9, 376, 2021.

[37] Liu, Y., Zhao, Y., Cheng, W., Zhang, T., *Targeted reclaiming cationic dyes from dyeing wastewater with a dithiocarbamate-functionalized material through selective adsorption and efficient desorption,* Journal of Colloid and Interface Science, 579, 766-777, 2020.

[38] Makhado, E., Pandey, S., Ramontja, J., *Microwave assisted synthesis of xanthan gum-cl-poly (acrylic acid) based-reduced graphene oxide hydrogel composite for adsorption of methylene blue and methyl violet from aqueous solution,* International Journal of Biological Macromolecules, 119, 255-269, 2018.

[39] Yamil, L.d.O., Georgin, J., Franco, D.S., Netto, M.S., Grassi, P., Piccilli, D.G., Oliveira, M.L., Dotto, G.L., *Powdered biosorbent from pecan pericarp (Carya illinoensis) as an efficient material to uptake methyl violet 2B from effluents in batch and column operations,* Advanced Powder Technology, 31, 2843-2852, 2020.

[40] Hamitouche, A., Haffas, M., Boudjemaa, A., Benammar, S., Sehailia, M., Bachari, K., *Efficient biosorption of methylene blue, malachite green and methyl violet organic pollutants on biomass derived from Anethum graveolens: An eco-benign approach for wastewater treatment,* Desalination Water Treatment, 5, 225-236, 2017.

[41] Li, P., Su, Y.-J., Wang, Y., Liu, B., Sun, L.-M., *Bioadsorption of methyl violet from aqueous solution onto Pu-erh tea powder,* Journal of Hazardous Materials, 179, 43-48, 2010.

[42] Priyantha, N., Lim, L.B., Tennakoon, D., Liaw, E.T., Ing, C.H., Liyandeniya, A.B., *Biosorption of cationic dyes on breadfruit (Artocarpus altilis) peel and core,* Applied Water Science, 8, 1-11, 2018.

[43] Chen, S., Zhang, J., Zhang, C., Yue, Q., Li, Y., Li, C., *Equilibrium and kinetic studies of methyl orange and methyl violet adsorption on activated carbon derived from Phragmites australis,* Desalination, 252, 149-156, 2010.