

PREPARATION OF CELLULOSE-BASED HYDROGEL DERIVED FROM TEA RESIDUE FOR THE ADSORPTION OF METHYLENE BLUE

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Cellulose was chemically extracted from tea residue and then a cellulose-based hydrogel was fabricated as an adsorbent to remove methylene blue from aqueous solutions. In this study, the cellulose-based hydrogel was prepared based on the copolymerization of acrylic acid (AA) on cellulose materials, with the addition of N,N'-methylene bis-acrylamide (MBA) as crosslinking agent, assisted by ammonium persulfate (APS) as initiator. The adsorption of the methylene blue (MB) solution onto the cross-linked cellulose-based hydrogel was studied by the static adsorption method under different conditions of pH, contact time, adsorbent dosage and initial methylene blue concentration. The results showed that for 50 mg/L of methylene blue solution as the initial concentration, the optimum pH was 10 and the adsorption efficiency was up to 98.89%. The analysis of the adsorption equilibrium isotherm strongly indicated that the adsorption by the hydrogel material was best fitted by the Langmuir model, with the maximum adsorption capacity of 41.67 mg/g. Furthermore, the cellulose-based hydrogel maintained high reusability and efficiency in methylene blue adsorption after desorption.

Keywords: tea residue, cellulose, hydrogels, adsorption, methylene blue

INTRODUCTION

In the process of national industrialization and modernization, environmental problems deserve special attention, not only in Vietnam, but worldwide. A system of environmental policies and institutions has been formulated and improved incrementally, to serve the environmental protection work towards sustainable development goals with higher efficiency. In spite of this, our environment continues to degrade rapidly – the quality of water resources has decreased enormously. One of the causes of the pollution is the textile industry in general, and dyes in particular.

Dyes may be defined as substances that provide colour when applied to a substrate, following an at least temporary alteration of the crystal structure of the coloured substances.¹ Dyes are classified according to their application and chemical structure. They are composed of a group of atoms known as chromophores, which

are responsible for the dye color. Methylene blue, also known as methyl thionium chloride, is a drug and a dye. The toxic effects of azo dyes may result from the direct action of the agent itself or of the aryl amine derivatives generated during reductive biotransformation of the azo bond.² The dyeing process involves a lot of water, so that waste water from textile dyeing is a huge pollutant around the world. Some dyes do not ever degrade in water. Others turn into harmful substances as they decompose.^{3,4} The ingestion of dye-contaminated water can cause serious problems due to their toxicity, highlighting the mutagenicity of its components, which strongly damage the health of living organisms.⁵

In recent years, various methods have been investigated to remove dyes from wastewaters, such as chemical methods, ion exchange, electrochemical, biological and membrane filtration techniques. Some of them are not

convenient because of high energy consumption and sludge production.^{1,6} Meanwhile, a novel idea for the removal of dyes from wastewater by the use of adsorbents based on agricultural by-products has received much attention. Biosorption is currently considered a promising technique for dye removal since it is an efficient, low-cost, simple method of treatment and can use a variety of waste materials from agriculture or other industries. Among bioadsorbent materials, hydrogels have gained attention due to their hydrophilic structure, which is capable of holding water in the three-dimensional network.^{7,8} Thus, biopolymer-based hydrogels are very attractive because they are capable of lowering the concentration of metal ions and other contaminants in wastewaters and can be prepared from widely available raw materials. A number of studies reported in the world and in Vietnam have demonstrated the ability of hydrogel materials to be used for environmental treatment and their potential for commercial utilization.

Cellulose ($C_6H_{10}O_5$)_n is a polysaccharide consisting of a linear chain of several hundreds to thousands of $\beta(1\rightarrow4)$ -linked D-glucose units, as shown in Figure 1. In general, cellulose is an important structural component of the primary cell wall of green plants, many forms of algae and oomycetes. Recently, has become an important chemical material resource.⁹ It is used to fabricate environmentally sustainable materials, with wide applications in tissue engineering, drug delivery, macroporous agriculture *etc.* Additionally, tea is the second most drank beverage in the world after water,¹⁰ especially in Asian countries, such as Vietnam, China and Japan, due to its health

benefits and distinct flavour. Green tea is a non-fermented tea and it contains higher amounts of polyphenols, caffeine, amino acids, vitamins, flavonoids and polysaccharides. According to the statistics of Food and Agriculture Organization of the United Nations (FAO), the overall production of tea beverages and instant tea results in high amounts of tea residue, which is discarded as waste. Hence, the utilization of tea residue would be of great significance, especially by turning it into a value-added product, and thus preventing loss of biological resources.¹¹

In this study, hydrogels were synthesized from cellulose, which was extracted from tea residue. The cellulose-based hydrogel was prepared by the copolymerization process of acrylic acid (AA), in the presence of N,N'-methylene bis-acrylamide (MBA) as cross-linking agent and ammonium persulfate (APS) as an initiator. Subsequently, the prepared cellulose-based hydrogel was used for the adsorption of methylene blue from aqueous solutions. The findings of this work will be of help in reducing environmental pollution. The production of the adsorbent hydrogels would utilise large quantities of waste, such as tea residue, for the treatment of dye-polluted wastewaters.

EXPERIMENTAL

Materials

Ammonium persulfate (APS), acrylic acid, methylene bis-acrylamide (MBA), methylene blue (MB), hydrochloric acid and sodium hydroxide were received from Xilong Scientific Co., Ltd, China.

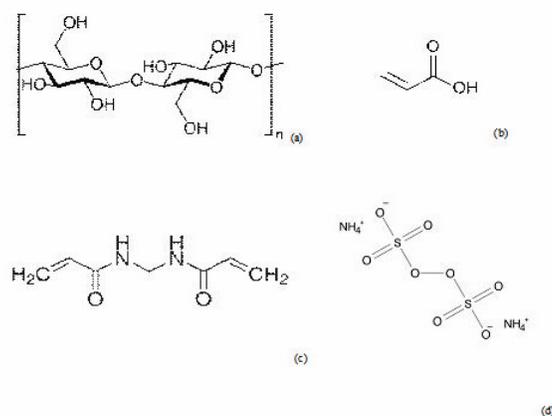


Figure 1: Chemical structures of cellulose (a), acrylic acid (AA) (b), N,N'-methylene bis-acrylamide (MBA) (c) and ammonium persulfate (APS) (d)

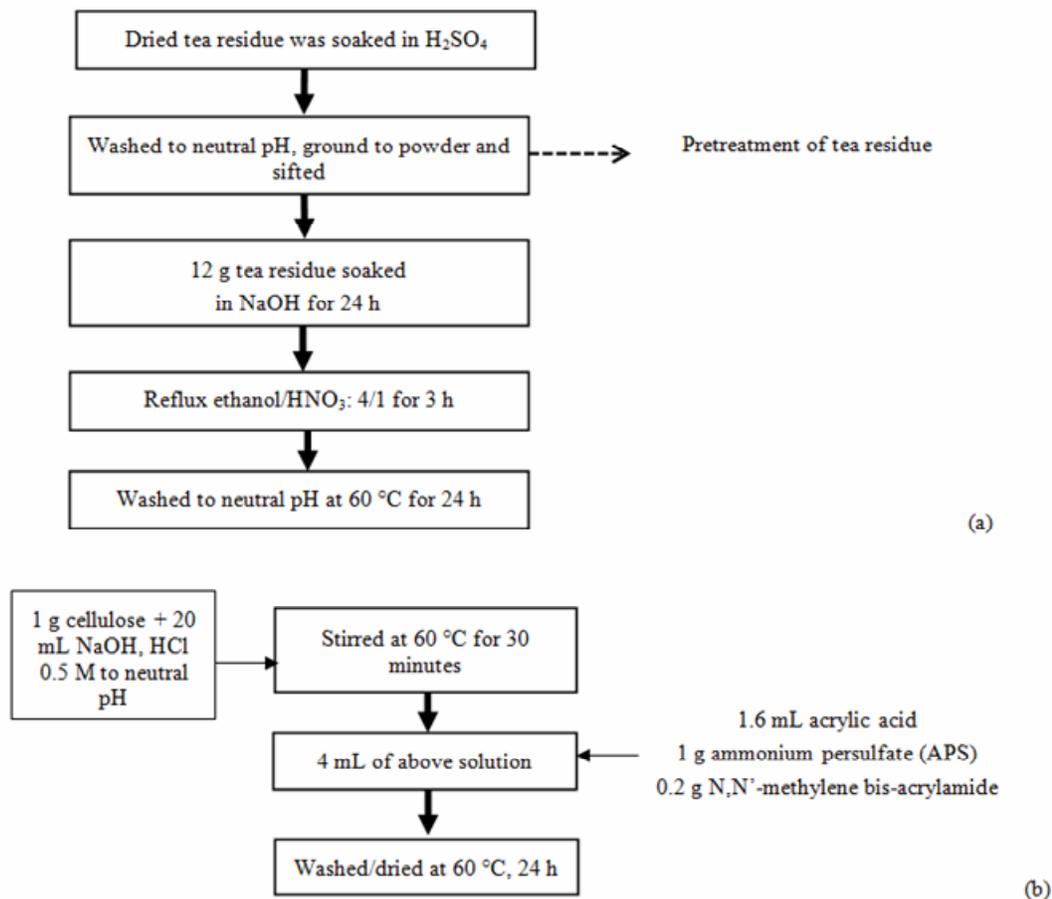


Figure 2: Cellulose extraction from tea residue (a) and preparation process of the cellulose-based hydrogel (b)

Methods

Extraction of cellulose from tea residue

The pretreatment of the tea residue is shown in Figure 2 (a). Briefly, the tea residue was collected from solid wastes from Longan province, Vietnam. About 10 g of the dried tea residue was soaked in 100 mL H_2SO_4 0.02 M at 70 °C for 3 h. It was then washed with distilled water to neutralise the pH and remove the debris. Finally, the tea residue was dried at 70 °C for 5 h, ground and sieved through a 60-mesh net.

In the next stages of the cellulose extraction process, 12 g of pretreated residue was soaked in distilled water for 12 h. Then, the tea residue was filtered and soaked in 300 mL NaOH 0.25 M at room temperature and washed to neutral pH again. After that, the tea residue was refluxed with an ethanol: HNO_3 (4:1 vol%) mixture for 3 h and the solvent was replaced every hour. The refluxed tea residue was filtered and washed with distilled water until neutral pH. The extracted cellulose was dried at 105 °C for 3 h to prepare for the following steps.¹²

Preparation of cellulose-based hydrogel

As shown in Figure 2 (b), 1 g of the extracted cellulose was added to 20 mL NaOH 0.5 M, then stirred at 60 °C for 30 minutes. After that, a HCl 0.5 M aqueous solution was added until the solution was neutral. For the formation of the cellulose-based hydrogel, about 1.6 mL of AA, 1 g of APS and 0.2 g MBA were added to about 4 mL of the above-mentioned solution and allowed to rest at 60 °C for 5 hours for the copolymerization process to take place. Then, the formed material was washed with distilled water and dried in a vacuum oven for 24 hours at 60 °C.¹³

Characterization of cellulose-based hydrogel

The morphology of the cellulose-based hydrogel was observed using a scanning electron microscope (JSM-5300LV, JEOL, Japan). Here, the hydrogel film was fractured in liquid nitrogen and freeze-dried for 24 h. Then, it was gold sputtered for the formation of a

conductive layer. Fourier transform infrared spectroscopy (FT-IR) was carried out after grinding dried hydrogel samples with potassium bromide (KBr), in the transmittance mode. The spectra were taken from 4000 to 400 cm^{-1} . The Brunauer-Emmett-Teller (BET) surface area of the cellulose-based hydrogel was analyzed by a Tristar II Surface Area and Porosity System (Shimazu). The hydrogel sample was dried under vacuum conditions over 24 h before nitrogen gas adsorption at 77 K. The swelling capacity (SC) of the hydrogel was measured at room temperature in this study.⁷ The value of SC was calculated for each sample by the following equation: $\text{SC} (\%) = (m - m_0) \times 100/m_0$, where m_0 is the dry weight and m is the weight of the hydrogel after 24-hour immersion in distilled water.

Methylene blue adsorption experiment

Effect of pH

Batch adsorption experiments using the cellulose-based hydrogel were conducted under various pH conditions. About 0.05 g of the cellulose-based hydrogel was placed in 20 mL of MB with the initial concentration of 50 mg/L, and shaken at room temperature, with constant speed at 180 rpm for 4 h. After that, the samples were filtered through a 5 μm filter paper and the concentration of the methylene blue solution was determined by a UV-Vis spectrophotometer at 664 nm.

Effect of contact time

Similar to the measurement of pH, contact time is one of the factors that greatly affect the adsorption capacity of a material. Herein, contact time experiments were carried out from 1 to 7 h, with the initial methylene blue concentration of 50 mg/L, at a pH of 10.

Effect of adsorbent dosage

The effect of material dosage was studied according to the above-described method to determine optimum adsorption conditions. The material dosage for the adsorption process was varied as follows: 0.02, 0.04, 0.06, 0.08, 0.1, 0.12 and 0.14 g. The other conditions were set to: contact time of 5 h, initial concentration of 50 mg/L and a pH of 10.

Effect of methylene blue concentration

The initial concentration of the contaminant solution greatly influenced the maximum adsorption capacity of the material. Initial concentrations of methylene blue of 25, 50, 100, 125 and 150 mg/L were used for a contact time of 5 h, at pH 10 and amount of the cellulose-based hydrogel of 0.04 g.

To study the adsorption equilibrium, the dried hydrogels were immersed into 20 mL of MB at room temperature. Upon the completion of the preset time intervals, the concentration of MB in the aqueous solution was measured. The adsorption capacity of the

hydrogel at time t (Q_t , mg/g) and the equilibrium adsorption capacity (Q_e , mg/g) were calculated according to the following equations, respectively:

$$Q_t = \frac{(C_0 - C_t) \cdot V}{m} \quad (1)$$

$$Q_e = \frac{(C_0 - C_e) \cdot V}{m} \quad (2)$$

where C_0 and C_t (mg/L) are the concentrations of the solution at the initial time and time t (h), respectively; C_e (mg/L) is the equilibrium concentration of methylene blue solution; V (L) is the volume of the contaminated solution and m (g) is the weight of the dried hydrogel.

The linear equation of the Langmuir model was used to correlate the adsorption equilibrium, as in the equation below:

$$\frac{C_e}{q_e} = \frac{1}{q_m \times K} + \frac{C_e}{q_m} \quad (3)$$

where q_m is the maximum adsorption capacity (mg/g) and K is the equilibrium adsorption constant related to the free energy of the adsorption test (L/mg).

Desorption and reusability of the hydrogel

MB-loaded hydrogels were treated with a HCl 0.1M aqueous solution at 180 rpm to desorb the contaminant. Subsequently, the adsorbent hydrogel was washed with distilled water and then reused for further adsorption processes. Adsorption/desorption cycles were successively conducted with fresh solutions until the hydrogel no longer showed methylene blue-adsorption capacity.

RESULTS AND DISCUSSION

Preparation of cellulose and cellulose-based hydrogel

The cellulose extracted from the tea residue was yellow coloured and darker after vacuum drying, as shown in Figure 3 (a, b). Judging by the yellow colour of the extracted cellulose, the pigments from the composition of the tea residue were not completely removed after the three-hour reflux process. The yield of the obtained cellulose was about 22%. As regards the cellulose-based hydrogel, it was prepared using APS as an initiator and MBA as a cross-linker *via* copolymerization of AA onto the cellulose extracted from the tea residue. In this method, the persulfate initiator decomposed under heating to produce sulphate anion radical. The radical abstracted the hydrogen from the hydroxyl groups of the cellulose to form initiating radicals on the polysaccharide chains. Thus, this persulfate-saccharide redox system provided active centres

on the substrate to radically initiate in the system, and the copolymer consisted of the crosslinking structure.¹⁴ The reaction scheme is illustrated in Figure 4. The hydrogel formed by this method had a white, moist surface after chemical

treatment (Fig. 3a). Then, the material was cut and dried to increase the adsorption contact area between the methylene blue and the hydrogel material, which, after drying in the vacuum oven, was dark yellow (Fig. 3b).

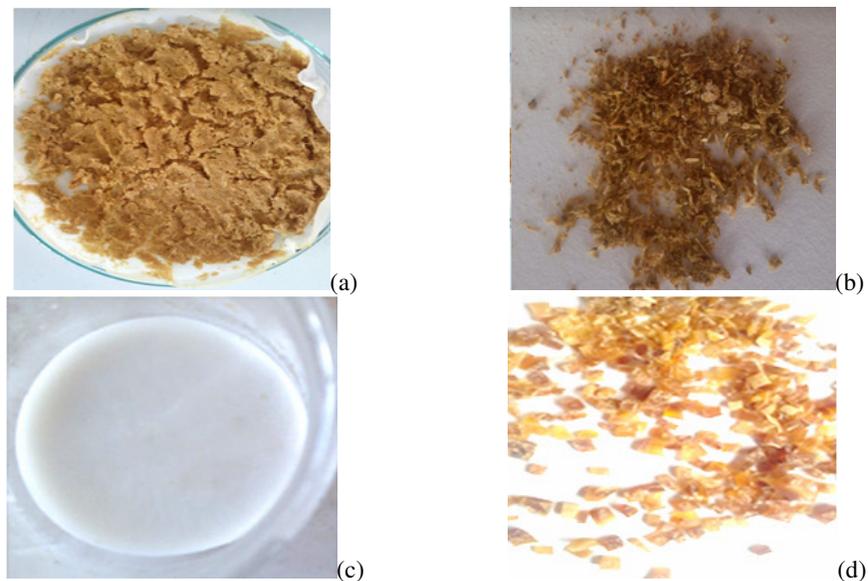


Figure 3: Cellulose extracted from tea residue before (a) and after (b) vacuum-drying; cellulose-based hydrogel before (c) and after drying in vacuum (d)

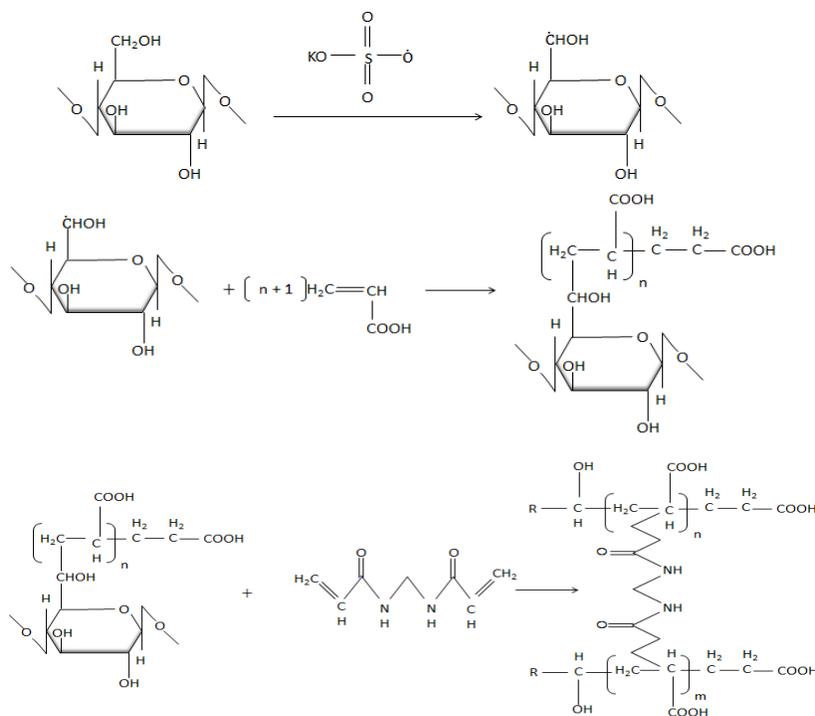


Figure 4: Reaction scheme of the preparation of the cellulose-based hydrogel in the presence of ammonium persulfate (APS), N,N'-methylene bis-acrylamide (MBA) and acrylic acid (AA)

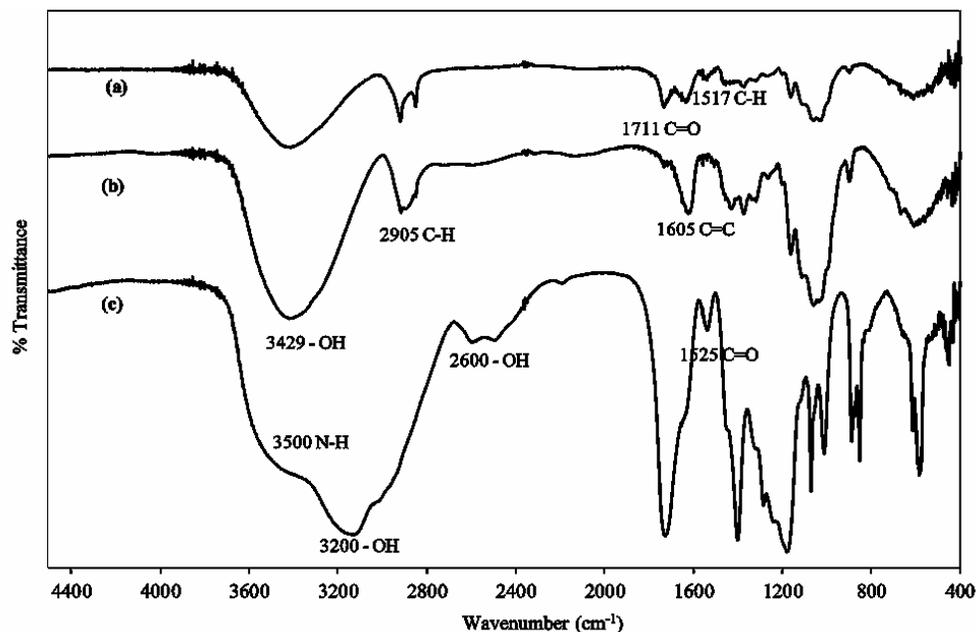


Figure 5: FT-IR spectra of extracted cellulose (a), tea residue (b), and cellulose-based hydrogel in the presence of the additives (c)

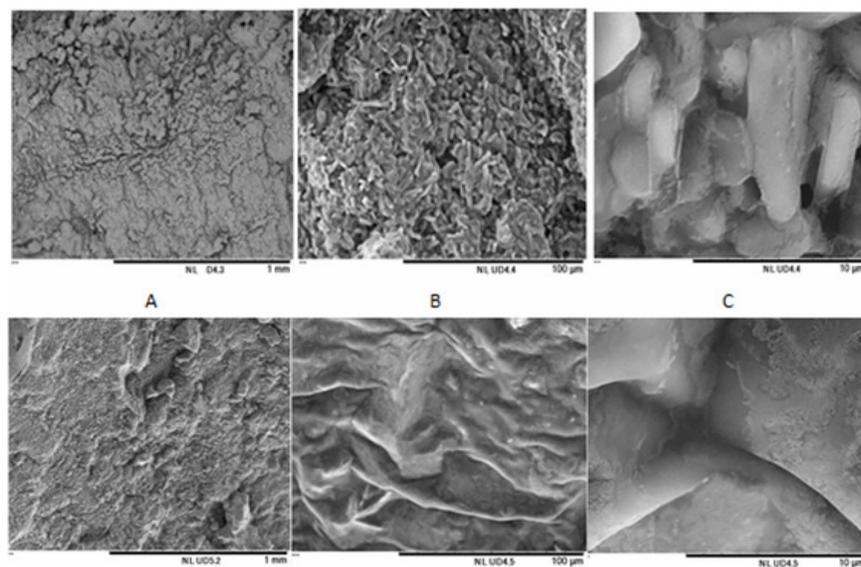


Figure 6: Surface (top row) and cross-sectional (bottom row) SEM images of the cellulose-based hydrogel at magnifications of $\times 100$ (A), $\times 1000$ (B) and $\times 10000$ (C)

The FT-IR spectra of the extracted cellulose and tea residue are displayed in Figure 5. According to it, the raw tea residue contained the characteristic functional groups of hydroxyl and C-H bending at the wavenumbers of 3429 cm⁻¹ and 2905 cm⁻¹. Moreover, typical peaks of hemicellulose and lignin components are noted at 1711 and 1517 cm⁻¹, which contributed to C=O and C-H bonds, respectively.¹⁵ However, as

shown in Figure 5 (b), the FT-IR spectrum of the extracted cellulose revealed a reduction in the content of hemicellulose and lignin by the decrease in the intensity of the peaks at 1711 cm⁻¹ and 1517 cm⁻¹. Also, the intensity of the peak at 1605 cm⁻¹ assigned to O-H stretching of the free water molecules was enhanced, suggesting the hydrophilicity of the prepared cellulose. Here, the spectra indicated that the amounts of the lignin

and hemicellulose components were reduced by the pretreatment process. The FT-IR spectra of the cross-linked cellulose-based hydrogel are displayed in Figure 5 (c). The hydrogel presented peaks attributed to the functional groups of C=O stretching at 1525 cm^{-1} for AA and N-H stretching in the wavenumber region of 3200 cm^{-1} for the (N-H-C=O) group stretching vibration of MBA.

SEM analysis indicated that the cellulose-based hydrogel exhibited many tear lines on the fractured surface (B). The SEM images in the top row show the surface of the cellulose-based hydrogel, displaying a rugged structure, a rough surface of the material, with the distribution of some bulges and a great number of holes on the surface. This indicates that MBA acted as a physical cross-linking agent to enhance the junctions and pores in the formed hydrogel. The SEM images in the bottom row of Figure 6 represent the cross-section of the formed hydrogel at different magnifications. They suggest that the hydrogel structure was porous, supplying a high surface area and large mass transfer channels, which increased the adsorption capacity of the hydrogel in aqueous solution. The BET result of the cellulose-based hydrogel material showed that the surface area of the material was quite modest: the dynamic correlation of the surface area of the material was not high, of $2.2\text{ m}^2/\text{g}$.

Swelling capacity is one of the most important physical properties of a hydrogel material, because it is determined by the amount of liquid that can be adsorbed. The higher the swelling capacity, the more durable the material is in an aqueous solution and the higher its adsorption capacity. The results obtained in this study indicated that the formed hydrogel had a great swelling capacity, with the maximum of 80.17% after 300 min immersion time, as shown in Figure 7.

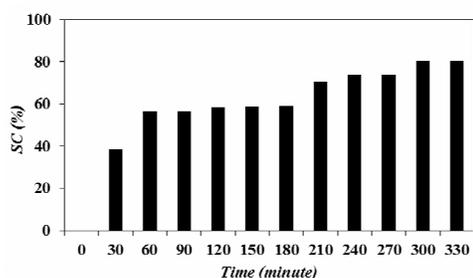


Figure 7: Swelling capacity of the cellulose-based hydrogel as a function of contact time

Methylene blue adsorption experiments

Effect of pH on the adsorption process

The pH is one of the most important factors that the adsorption capacity of a hydrogel material. Methylene blue has ammonium groups acting as cationic substances in a wide range of pHs. Thus, the changes of the adsorption capacity could be due to differences in the active sites on hydrogels at a specific pH. At a pH lower than 7, the surface of hydrogels would be positively charged. Therefore, this would limit the approach of positively charged methylene blue because of electrostatic repulsion. An increment in the pH of the solution leads to increased adsorption capacity of the hydrogels for methylene blue, as the number of positively charged active sites on the hydrogel decreases. The maximum adsorption capacity of the hydrogel was observed at a pH higher than 7, suggesting an enhanced number of negatively charged active sites on the hydrogel.¹⁶

Figure 8 shows the variation in methylene blue adsorption efficiency of the hydrogel material at different pH values. The results suggest that at pH = 10, the removal capacity of MB was the highest (H = 98.99%, Q = 17.74 mg/g), and therefore this would be the optimum pH level for the adsorption.

Effect of contact time on the adsorption process

The effect of contact time on the adsorption of methylene blue was also investigated. It may be noted that the adsorption rate increases rapidly in the first 5 h, because the active sites were vacant at the beginning of the adsorption process. After that, the rate of methylene blue uptake decreased owing to the occupation of these active sites. The optimum removal efficiency was achieved after about 5 h contact time with the hydrogel material. The results in Figure 9 showed that after 5 h, the material reached the maximum adsorption capacity (Q = 19.03 mg/g, H = 97.22%).

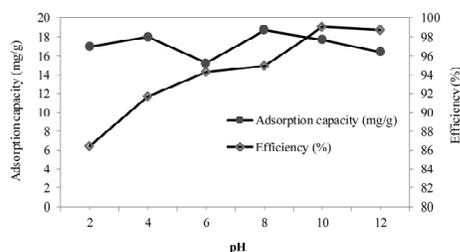


Figure 8: Effect of pH on the adsorption process

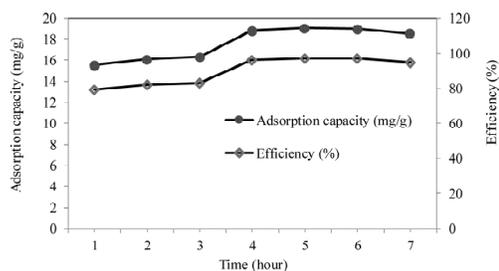


Figure 9: Effect of contact time on the adsorption process

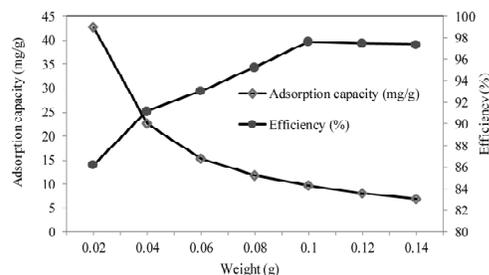


Figure 10: Effect of adsorbent dosage on the adsorption process

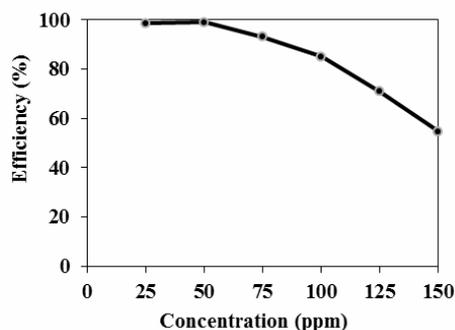


Figure 11: Effect of methylene blue concentration on the adsorption process

Effect of adsorbent dose on the adsorption process

The removal efficiency of MB increased when increasing the adsorbent dose, which meant the number of the adsorbent active sites increased, albeit slowly. Figure 10 illustrates a continuous increase in the adsorption efficiency with a rising weight from 0.02 g to 0.1 g. When using 0.02 g hydrogel, the efficiency was of 86.22%, which increased to 91.21% when using 0.04 g of adsorbent. The optimum dosage of the adsorbent material was considered that of 0.04 g, when high efficiency and adsorption capacity were observed ($Q = 22.58 \text{ mg/g}$, $H = 91.21\%$).

Effect of methylene blue concentration on the adsorption process

Figure 11 shows the evolution of the adsorption efficiency with a rising initial concentration of methylene blue. At first, the adsorption efficiency is quite high, because the adsorption sites of the gel are empty initially. With the increment of the initial concentration of MD from 25 to 50 mg/L, the removal capacity of the cellulose-based hydrogel increased slightly, since more methylene blue ions were available to react with the active sites of the hydrogel. However, at very high initial concentrations, the

amount of the adsorbed methylene blue remained constant as the active sites of the hydrogel reached saturation and as a result, the adsorption efficiency had a decreasing trend. Therefore, 50 mg/L was considered as the optimum initial concentration of methylene blue to achieve highly efficient adsorption by the prepared hydrogel ($Q = 24.72 \text{ mg/g}$, $H = 98.89\%$).

The Langmuir and Freundlich constants were calculated and are listed in Table 1. As shown, the correlation coefficient obtained by the Langmuir model ($R^2 = 0.9979$) was higher than that of the Freundlich model ($R^2 = 0.7725$), which strongly indicates that the Langmuir isotherm model is more suitable to describe the adsorption behaviour of the hydrogel material from the current experiment, suggesting that a monolayer of methylene blue covered the adsorbent (Fig. 12).

A number of reports are known regarding the adsorption of MB using cellulose-based adsorbents with high removal efficiencies.^{3,4,8} However, MB desorption experiments on the adsorbent have not been mentioned in these studies. Therefore, in this research, the reusability of the hydrogel material was evaluated. The results indicated that the potential reusability of the prepared material was very high, which means

it could be used for commercial purposes.

Table 1
Sorption isotherm correlation coefficients and constants

Langmuir	Q_m (mg/g)	41.841
	K_L (L/mg)	3.3662
	R^2	0.9979
Freundlich	K_F	21.414
	1/n	0.2009
	R^2	0.7725

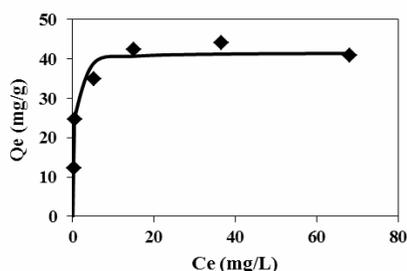


Figure 12: Isotherm for methylene blue adsorption by the hydrogel

To be considered as a promising adsorbent, the materials should present high adsorption capacity for contaminants and high reusability for a number of cycles. As observed in our adsorption-desorption experiments, the pH of the eluted solution plays a great role in the desorption behaviour of the hydrogel. The results suggested using acidic aqueous solutions to desorb the MB-loaded hydrogel due to the exchange between the loaded MB on the active sites of the hydrogel and the H^+ ions. During five cycles of utilization, the removal capacity of the hydrogel did not differ significantly (Fig. 13). Judging by the high adsorption capacity and good recyclability of the hydrogel, it can be considered a good candidate for practical applications in the removal of pollutants from effluents.

CONCLUSION

In the present study, a cellulose-based hydrogel has been successfully synthesized from tea residue, and the prepared material demonstrated the ability to adsorb methylene blue from aqueous solutions. According to experimental results, the following conditions were established as optimum for the adsorption: pH 10, initial concentration of methylene blue in the solution of 50 mg/L, contact time of 5 hours and 0.04 g of hydrogel dose. The efficiency of the

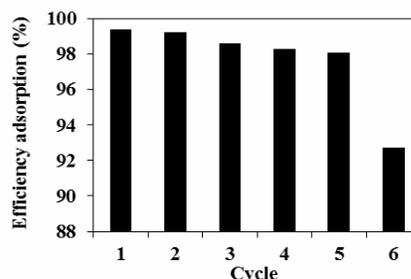


Figure 13: Reusability of the cellulose-based hydrogel for methylene blue adsorption

adsorption process under these conditions was of 98.89%. The adsorption equilibrium data fitted well the Langmuir isotherm model, with the maximum adsorption capacity of 41.67 mg/g. Furthermore, the developed hydrogel presented good reusability and high efficiency in dye adsorption.

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