Evaluate to remove azo dye Chrysoidine Y of activated carbon material produced from bamboo leaves

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ABSTRACT

Activated carbons (ACs) are well known as the most commonly used adsorbent in water and wastewater treatment. They have many advantage characteristics such as high surface area, large pores and diverse surface functional groups with the high stability of chemical, mechanical and thermal. The aims of this study are to fabricate and evaluate the ability to remove the azo dye Chrysoidine Y in the water of three activated carbon materials from bamboo leaves, including AC30 (650°C/30 minutes); AC45 (650°C/45 minutes) and AC60 (650°C/60 minutes). The material characteristics determined by SEM, EDX, FTIR and BET methods, etc. showed that all three samples had high C content (over 72%) and appeared functional groups with the capacity of removing azo dye as C = C; C - O - C, O - H. The FTIR results indicated that three samples have adsorption spectra from 400÷4000 cm⁻¹ with characteristic bonds such as C = O, O - H, C - C, C = C. When 20 ml of a solution containing 0.25; 0.5; 1 and 2 g of AC30, AC45 and AC60 materials were added the azo dye volume varied from 2, 3, 4 and 5 ml of Chrysoidine Y, the lowest and highest adsorption capacity were recorded of 133.64 and 361.2 mg/g, respectively. With the highest BET surface area up to 108,9202 m²/g, the AC60 material sample achieved a maximum efficiency of 100% at a reaction time of 30 minutes with a Chrysoidine dye/distilled water volume ratio of 2/18 (ml), pH 9, adsorbent content of 0.25 g. The results also showed that the activated carbon from bamboo leaves is a potential sorbent material in removing the azo dye in water.

1. Introduction

Azo dyes are widely used in various industries such as textile, leather, plastic, paper and ink production, accounting for more than 60% of total dyes (Benkhaya et al., 2020). Azo dyes are characterized by the presence of one or
more azo functional groups $(R_1 - N = N - R_2)$, where $R_1$ and $R_2$ are aromatic groups, can be substituted by some combination of functional groups such as amino ($-\text{NH}_2$), chlorine ($-\text{Cl}$), hydroxyl ($-\text{OH}$), methyl ($-\text{CH}_3$), nitro ($-\text{NO}_2$), sulphonlic acid and sodium salt ($-\text{SO}_3\text{Na}$) (Benkhaya et al., 2020). However, they are also a group of serious water pollutants and currently, there is great interest in finding ways to completely remove these substances from water. Most dyes are toxic, carcinogenic, or mutagenic (Hao et al., 2015) and can pose health hazards, such as chromosomal damage (Aboel-Zahab et al., 1997); affecting human eyes and skin, damage to internal organs such as the liver and kidneys (Ahmad et al., 2020); disrupts photosynthesis in water bodies (Oveisi et al., 2018), etc. Therefore, it is very necessary to treat textile dyeing wastewater or to find new materials to adsorb or assist in removing azo dyes from wastewater. Several techniques have been studied to remove azo dyes from wastewater (Srinivasan and Viraraghavan, 2010), including chemical, physical and biological treatments or a combination of these processes such as adsorption: (Kył et al., 2020), hydrogen peroxide (Krishna et al., 2016), biological treatment (Kornaros and Lyberatos, 2006), membrane filtration (Lee et al., 2006), etc. Among the physical processes, the adsorption method is considered as an efficient separation with regard to low cost, simple design, ease of operation and sensitivity to hazardous substances (Abd-Elhamid et al., 2020).

Activated carbons (ACs) are well known as the most commonly used adsorbent in water and wastewater treatment. ACs have the advantages of high surface area, large pores and diverse surface functional groups with the high stability of chemical, mechanical and thermal (Elmaguana et al., 2020). ACs are used in many fields such as removal of organic pollutants, heavy metals, medical, catalysis, electrode materials in electrochemical devices (Amirza et al., 2017), removal of azo dyes (Khamis et al., 2020), etc. In recent years, ACs adsorption technique has been widely applied in water treatment due to its fast adsorption kinetics, simple design, economic cost, high removal efficiency and insensitivity to toxic substances. ACs are fabricated from a variety of raw materials, including industrial waste (Dias et al., 2019), agricultural by-products (Vu et al., 2017), livestock and poultry manure (Zhu et al., 2018), vermicompost mud (Yang et al., 2016).

Bamboo is a large woody plant in the family Bambusoideae which includes about 1250 species in 75 genera over the world (Fei et al., 2016). Vietnam is the 4th country in the world in terms of bamboo growing area and bamboo production, mainly grown in Lam Dong and Da Lat. Nowadays, the study of low-cost activated carbon production from bamboo is increasingly interested. However, there are not many publications on the use of ACs material from bamboo leaves to adsorb azo dyes from industrial wastewater. Therefore, in this study, the activated carbon fabricated from bamboo leaves at 650°C for 30, 45 and 60 minutes was used to evaluate the ability to remove Chrysoidine Y azo dye from an aqueous solution.

2. Methodology

2.1. Preparation and characterization of activated carbon material

Fresh bamboo leaves were collected at Lang street, Thach That district, Hanoi city and transferred to the laboratory, washed and naturally dried at room temperature for 24 hours. Bamboo leaves were then chopped and soaked with 5% $\text{H}_3\text{PO}_4$ solution for 1÷2 hours and taken out to drain. The preparation steps were conducted to comply with the procedure by Tran et al., (2021) such as: activating them at a temperature of $225\pm230^\circ\text{C}/4$ h, down the temperature of the oven to 150°C and taking them out, washed with the distilled water and drain naturally. Then, the activated bamboo leaves were dried at 60°C for 12 hours. Finally, the carbonized bamboo leaves were transferred to activate in the UAF furnace at a temperature of 650°C for 30, 45 and 60 minutes, respectively. The material samples were dried at 105°C for 12 h, finely ground and stored in dark bottles. The three material samples will be encoded as AC30, AC45 and AC60 and used for further experiments.

Material characteristics were determined by techniques: scanning electron microscope (SEM - Scanning Electron Microscope), energy-dispersive X-ray spectroscopy (EDX - Energy-dispersive X-ray) and functional groups on the material surface were determined by Fourier...
transform infrared spectroscopy (FTIR) TENSOR II and the specific surface area (BET - Brunauer-Emmett-Teller) and Particle Size Distribution (PSD) of the material was determined at the Institute of Tropical Technology - Vietnam Academy of Science and Technology.

2.2. Experimental design and evaluate the adsorption capacity of the material

Weigh 0.1 g Chrysoidine Y and dissolve in 100 ml alcohol, shake well and store in a dark bottle as a stock solution for further experiments. The experimental procedure was conducted to comply by Tran et al., 2021 follow as:

The first step, conduct experiments to select the optimal ratio of Vazo dye/distilled water: weigh 0.5 g of activated carbon and put it into 20 ml of a solution containing azo dye Chysoidine Y and distilled water according to the ratio of Vazo dye/distilled water = 2:18; 3:17; 4:16; 5:15 and no pH adjustment. Then, shake the samples with a shaker at 150 rpm for 30, 60, 90 and 120 minutes and measure the pH of these samples. Filter the sample with blue-band filter paper Ø = 0.45 µm and analyze the concentration of azo dye in the sample by HPLC instrument at wavelength 415 nm at the Institute of Chemistry - Material, Institute of Military Science and Technology. The ratio of Vazo dye/distilled water with the highest adsorption efficiency was selected to evaluate for the next experiments.

Then weigh 0.25, 0.5, 1 and 2 g of three material samples into each beaker containing 20 ml of solution with the selected optimum ratio and continue to perform experiments at 30, 60, 90 and 120 min. The adsorption capacity of Qe (mg/g) and the treatment efficiency (H%) at equilibrium were determined by the equation suggested by Tran et al., 2020 such as:

\[ Qe = (Co-Ce).V/m \text{ (mg/g)} \]  
\[ H (%) = (Co-Ce)/Co.100 (%) \]

Where: Co - initial concentration (mg/l); Ce - equilibrium concentration (mg/l); V - volume of the dye solution (ml); m - mass of the sorbent added (g).

2.3. Statistical data

The collected data in this study were processed by following software such as GraphPad 6, Origin 2019b with a statistical significance of ρ <0.05.

3. Results and discussion

3.1. Structural characterization of fabricated materials

The structural characterization of the three activated carbon samples in this study is shown in Table 1 and Figures 1 and 2.

<table>
<thead>
<tr>
<th>Samples</th>
<th>Time and temperature</th>
<th>BET surface area (m²/g)</th>
<th>Content of surface elements (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>C (%)</td>
</tr>
<tr>
<td>AC30</td>
<td>650°C/30 m</td>
<td>25.2337</td>
<td>72.45</td>
</tr>
<tr>
<td>AC45</td>
<td>650°C/45 m</td>
<td>44.6221</td>
<td>74.20</td>
</tr>
<tr>
<td>AC60</td>
<td>650°C/60 m</td>
<td>108.9202</td>
<td>80.18</td>
</tr>
</tbody>
</table>

Table 1. Physical and chemical characteristic of three activated carbon samples.

Figure 1. SEM images of three activated carbon samples: a) AC30; b) AC45 and c) AC60.
All the samples AC60, AC45 and AC30 have content of C rather high with values of 80.18; 74.2 and 72.45%, respectively. The particle size distribution ranged from 17.4889 µm (AC30 sample) to 23.497 µm (AC45 sample) and the specific surface area (BET) of AC60 sample was recorded up to 108.9202 m²/g and PSD value of 18.7502 µm. All three material samples have a heterogeneous porous structure, the AC60 sample had a more porous, diameter of porous larger. This result is consistent with some previously published studies (Cui et al., 2015; Vu et al., 2017). It can be seen that the material with a large pore diameter reduces the surface area (Vu et al., 2017) and improves the adsorption capacity for azo dyes. Compared with raw materials, activated carbon materials have stronger adsorption capacity through the rough surface with tiny channels and porous structures (Cui et al., 2015).

Similarly, the FTIR spectrum in Figure 2 indicated that the time and temperature of the activation process affect the material properties and the formation of functional groups on the material surface (Pavia et al., 2013). All three samples have adsorption spectra from 400 to 4000 cm⁻¹ with characteristic bonds such as C = O, O - H, C - C, C = C, etc. Moreover, according to Pavia et al., (2013), FTIR spectra of three activated carbon samples in this study are different from many others, because they contain spectra around 2350 cm⁻¹ representing the CO₂ group. The activation time ranging from 30, 45 to 60 will affect to form or loss of the functional groups on the surface material and the intensity of the peaks also changes. The C = O bond in the form of a ketone group (1725±1705 cm⁻¹) is only observed in sample AC30. This feature shows that the AC30 sample contains more functional groups on the surface of the material, so the purity will be lower than the two samples of AC45 and AC60 left. This result leads to more dispersion and reduced adsorption efficiency of AC30 sample.

3.2. Evaluation of the adsorption capacity of three activated carbon materials for removing azo dye

3.2.1. Effect of the original azo dye content and reaction time on the adsorption capacity of the material

To study the effect of original azo dye content and reaction time on adsorption capacity, 0.1 g Chrysoidine Y was diluted in 100 ml alcohol and analyzed within the original concentration by HPLC instrument at wavelength 415 nm. The analysis result recorded the real concentration in the stock solution of 90.3 mg/l. As the volume of azo dye added to the sample varied from 2, 3, 4 to 5 ml and the reaction time increased from 30÷120 min, the azo dye removal capacity also increased accordingly (Table 2).

All samples recorded pH values changed from 9÷9.4. The AC60 sample recorded the highest adsorption efficiency when adding 2 ml of Chrysoidine Y azo dye, at all 4 time periods, the adsorption efficiency was recorded at 100% corresponding to the maximum adsorption capacity (Qe) of 361.2 mg/g. When the volume of azo dye was increased to 3, 4, 5 ml, the lowest adsorption efficiency was recorded at 80% (corresponding to Qe = 288.96 mg/g) with 5 ml in 30 min reaction time and reached the maximum value (100%) when the reaction time

![Figure 2. FTIR spectrum of three activated carbon samples: a) AC30; b) AC45 and c) AC60.](image-url)
Table 2. The adsorption capacity $Q_e$ (mg/g) of Chrysoidine Y azo dye by three activated carbon materials.

<table>
<thead>
<tr>
<th>Types of materials</th>
<th>Vazo dye (ml)</th>
<th>Material mass (g)</th>
<th>$Q_e$ (mg/g)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>30 m</td>
</tr>
<tr>
<td>AC30</td>
<td>2</td>
<td>0.5</td>
<td>352.17</td>
</tr>
<tr>
<td>AC45</td>
<td>2</td>
<td>0.5</td>
<td>355.42</td>
</tr>
<tr>
<td>AC60</td>
<td>2</td>
<td>0.5</td>
<td>361.20</td>
</tr>
<tr>
<td>AC30</td>
<td>3</td>
<td>0.5</td>
<td>339.53</td>
</tr>
<tr>
<td>AC45</td>
<td>3</td>
<td>0.5</td>
<td>344.58</td>
</tr>
<tr>
<td>AC60</td>
<td>3</td>
<td>0.5</td>
<td>356.14</td>
</tr>
<tr>
<td>AC30</td>
<td>4</td>
<td>0.5</td>
<td>133.64</td>
</tr>
<tr>
<td>AC45</td>
<td>4</td>
<td>0.5</td>
<td>227.56</td>
</tr>
<tr>
<td>AC60</td>
<td>4</td>
<td>0.5</td>
<td>307.02</td>
</tr>
<tr>
<td>AC30</td>
<td>5</td>
<td>0.5</td>
<td>133.64</td>
</tr>
<tr>
<td>AC45</td>
<td>5</td>
<td>0.5</td>
<td>216.72</td>
</tr>
<tr>
<td>AC60</td>
<td>5</td>
<td>0.5</td>
<td>288.96</td>
</tr>
</tbody>
</table>

was increased to 120 min. For AC45 sample, the highest efficiency reached 100% ($Q_e = 361.2$ mg/g) at a reaction time of 120 min when 2 and 3 ml of Chrysoidine Y azo dye were added. As the volume of azo dye increased to 4 and 5 ml, respectively, the adsorption efficiency varied with the lowest value of 60% (with 5 ml for 30 minutes, $Q_e = 216.72$ mg/g) and the highest value was 96.7% (with 4 ml for 90 minutes, $Q_e = 349.28$ mg/g). In three materials, the AC30 sample has the lowest removal ability for azo dye, the lowest adsorption efficiency is only 37% (with 4 and 5 ml in 30 minutes, $Q_e = 133.64$ mg/g) and the highest is 100% (with 2 and 3 ml for 120 min). From the survey results, it was found that the ratio of Chrysoidine Y/distilled water $= 2/18$ was the most optimal and was selected for further experiments.

3.2.2. Effect of the adsorbent content and reaction time to the removal of azo dye

From the results obtained in section 3.2.1, a volume ratio of azo dye Chrysoidine Y/distilled water (ml) $= 2:18$ was chosen for the subsequent experiments. The experimental results are shown in Figure 3. When increasing the amount of adsorbent from 0.25; 0.5; 1÷2 grams, the adsorption efficiency (H%) for azo dyes also increased. The AC60 sample achieved the adsorption efficiency of 100% at the reaction time of 120 min with all 4 ratios of the amount of adsorbent 0.25, 0.5, 1 and 2 grams. With the different periods, the adsorption efficiency varied from 80% (with 0.25 g at 30 min) to 97.7% (with 0.25 g at 90 min) and reached 100% as the amount of absorbent material increased to 2 g. The AC30 sample has a rather low removal efficiency, the lowest efficiency was 37% (with 0.25 g at 30 minutes) and only reached 100% when the absorbent mass was 1 g at 90 minutes. Similarly, the AC45 sample recorded the removal efficiency of 60% (with 0.25 g at 30 min) and this value also reached 100% with 1 g at 90 min. Thus, only AC60 sample can remove 100% of Chrysoidine Y azo dye when adding 0.5 g of adsorbent material in 90 minutes or 1 g in 30 minutes, the remaining two samples only achieved the maximum removal efficiency when the reaction time increased to 90÷120 minutes and the absorbent material content increased from 1 g to 2 g.

Azo dyes are a group of dyes represented by a functional group (-N = N-) and the removal of this group from textile dyeing wastewater is a very difficult task. Our experiments confirm previous results (Kaya and Uzun, 2020; Chan et al., 2008; Hameed and El-Khaiary, 2008). Kaya and Uzun (2020) used the activated carbon materials from pinecones, walnut shells and hazelnut shells synthesized at different temperature ranges from 400÷700°C for 60 min to remove Alizarin yellow GG azo dye from an aqueous solution. The results showed that when 20 ppm Alizarin yellow GG and 8 g/l of absorbent...
material were added into the solution at pH 3, the activated carbon sample from walnut shell had reached the highest adsorption efficiency of 82% with the largest surface area up to 259.74 m²/g. Similarly, Chan et al., (2008) showed that dyes with smaller molecular sizes are more easily absorbed into carbon materials and the removal efficiency of dye depends on the presence of functional groups on the surface of the materials. The results showed that Acid Blue25 (AB25) dye with a smaller molecular size is easier to absorb into carbon materials than Acid Yellow 117 (AY117) dye with a larger size. The material characterization also indicated that the FTIR spectrum of the activated carbon sample appeared in functional groups such as C = C, O - H, C - O - C representing peaks at 1650, 3450 and 1120 cm⁻¹. These groups play an important role in the dye adsorption process. The surface structure and porous size of the material also play an important role in the dye adsorption and removal process. Hameed and El-Khaiary (2008) demonstrated that the activated carbon samples with high BET specific surface area and large porous size will have the adsorption capacity with pollutants also increased accordingly.

The reaction time and the initial adsorbent concentration during the batch adsorption process are important factors in determining the adsorption capacity. The longer the adsorption time, the higher the adsorption efficiency (Saeed et al., 2010; Laskar and Kumar, 2018). In the current study, when the amount of the initially activated carbon increased from 0.25 g, the AC30 sample also recorded the maximum removal efficiency of 100% after only 30 minutes of reaction time, two samples AC60 and AC45 recorded this efficiency with 0.25 g and 2 g, respectively, after only 30 minutes of reaction time. Laskar et al., (2018) showed that the removal efficiency of azo Brilliant green (BG) by the activated carbon synthesized from Bambusa Tulda bamboo reached a maximum value of 98% after 60 minutes with a weight of adsorbent material of 10 g/l. The BG removal rate was reduced to 83.74% when the adsorbent material concentration was reduced to 50 mg/l. Moreover, types of activated carbon different also

Figure 3. The adsorption efficiency (H%) of Chrysoidine azo dye from aqueous solution when adsorbed with 0.25; 0.5; 1 and 2 grams activated carbon material.
have different adsorption efficiency and different azo dyes are also removed at different rates. Laskar et al., (2008) also demonstrated that the removal rate of BG azo dye was reduced from 136±72 mg/l after adsorption by activated carbon modified with sodium carbonate. With another azo dye of crystal violet (CV), Saeed et al., (2010) used the carbonaceous material from pomelo peel and the result showed that the adsorption equilibrium quickly reached 96% after 60 minutes. The maximum adsorption capacity gradually increased from 60.42±254.16 mg/g when the adsorbent content also increased from 25 mg/l to 1 g/l and the amount of CV added to the solution increased accordingly from 10÷600 mg/l (Saeed et al., 2010).

The effect of experimental conditions such as pyrolysis time and temperature, pH and reaction time, etc. on the azo dye removal processing is obvious. According to Mahdi et al., (2017), the pyrolysis time will determine the material recovery yield and affect the structure of cellulose and hemicellulose, which leads to the release of organic molecules. The formation of functional groups (C = O, C-O-C and C-O) will determine the ability to remove the azo dye and improve the surface characteristics of the material (surface area and porous volume). The efficiency of adsorption of pollutants depends on the polar groups on the surface of the carbonate material, so if there are many hydroxyl groups, carboxyl groups and functional groups such as -OH, CH, C=O, etc., on the surface material, the absorption capacity will have a strong influence (Hirata et al., 2020). The conditions of the material synthesis environment, experimental factors and the isothermal adsorption mechanism will greatly affect the complete removal capacity of the materials (Macedo et al., 2006). It can be seen that the origin and nature of materials for the synthesis of carbonaceous materials have a great influence on the material properties, determining the treatment efficiency as well as the pollutant removal processing. The current results showed that the material synthesis conditions (pyrolysis time and temperature) and the volume ratio of azo dye added to the solution are suitable for the application of removing the azo dye in textile dyeing wastewater.

4. Conclusion

This study investigated the removal capacity of azo dye Chrysoidine Y by three types of activated carbon prepared from bamboo leaves, AC30 (650°C/30 min); AC45 (650°C/45 min) and AC60 (650°C/60 min). The material characteristic has a relatively high C content (AC30 is 72.45%; AC45 is 74.30% and AC60 is 80.18%) and contains the specific bonds for azo dye removal such as C = C; C - O - C, - OH. The results also showed that the AC60 material sample had the maximum adsorption efficiency of 100% after 30 minutes with the azo dye/distilled water volume ratio of 2/18 (mg/l), pH 9 and the amount of adsorbent 0.25 g. The results show that the activated carbon from bamboo leaves is a potential material resource for removing the azo dye in water. More research is needed to have a deeper investigation into the adsorption mechanism, wastewater properties, desorption/adsorption, etc. so that it can be applied in production plants or actual wastewater samples.

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Author contributions

Huong Thu Thi Tran - contributes to the idea, data acquisition, analysis and writes the manuscript; Ha Kim Thi Tran - contributes to collecting the data.

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