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Synthesis of Activated Carbon from Polyethylene Terephthalate (PET) Plastic Waste and Its Application for Removal of Organic Dyes from Water

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ABSTRACT

Synthetic plastics are often considered to be materials that cannot be broken down by natural processes. One such plastic, polyethylene terephthalate (PET), is commonly used in everyday items but when these products are discarded, they can cause serious harm to the environment and human health. In this study, PET plastic waste was used to create activated carbon using a physical activation process that involved using CO_2 gas. The researchers investigated the effects of different temperatures, carbonization, and activation times on the resulting activated carbon's surface area. The activated carbon was then analyzed using scanning electron microscopy (SEM), X-ray diffraction (XRD), FTIR, and BET. The activated carbon created from PET plastic waste showed excellent absorption properties for methylene blue in aqueous solutions across a wide range of pH levels. By creating activated carbon from plastic waste, not only are environmental issues addressed, but high-value activated carbon is produced for environmental remediation purposes.

Keywords: Plastic waste; Environmental treatment; Activated carbon; Waste utilization; Polyethylene terephthalate (PET)

1. Introduction

Plastic is widely used in all aspects of daily life and production, such as packaging, household items, construction, transportation, electrical industry, electronics, and other applications. In 2017, over 348 million tons of plastic were produced worldwide, and it is predicted to quadruple by 2050 [1]. Since its introduction into civilian production and application in the 1950s, approximately 6.3 billion tons

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ARTICLE INFO
Received: 18 April 2023 | Revised: 26 April 2023 | Accepted: 27 April 2023 | Published Online: 12 May 2023
DOI: https://doi.org/10.30564/nmms.v5i1.5663

CITATION
Pham, T.H.T., 2023. Synthesis of Activated Carbon from Polyethylene Terephthalate (PET) Plastic Waste and Its Application for Removal of Organic Dyes from Water. Non-Metallic Material Science. 5(1): 27-37. DOI: https://doi.org/10.30564/nmms.v5i1.5663

of plastic have been discarded, of which about 79% were buried and disposed of in the environment [2]. Plastic waste is very difficult to decompose in the natural environment. Each type of plastic has a different number of years to decompose, with a very long time, hundreds of years, and sometimes even thousands of years. Plastic waste has caused many negative impacts on the ecological environment and human health, therefore finding a solution to plastic waste disposal is one of the most urgent issues.

Currently, most commercial plastic recycling plants mainly use mechanical processing and recycling methods. In Europe, over 5 million tons of plastic waste are recycled by mechanical methods, while only about 50,000 tons are processed and recycled by chemical methods [3]. Mechanical processing methods are often complex and require separate waste collection lines, leading to relatively high costs. In addition, chemical recycling and processing technologies are considered to be economically viable and capable of achieving complete recycling. Many scientists around the world have recently focused on developing technology to recycle plastic waste into carbon-based materials such as graphene, carbon nanotubes, and especially activated carbon [4-9]. Physical activation is a widely used technology for recycling plastic waste into activated carbon [10-13]. Qiao's group used hot steam to activate PVC in the production of activated carbon [4]. PET plastic waste is also commonly used as raw material for the production of activated carbon [7,14-17]. First, PET is carbonized at a temperature of 700 °C in a nitrogen gas environment with a carbon yield of about 20% [5]. The physical activation method also uses CO_2 to activate carbon from PET plastic. Almazán-Almazán and colleagues have successfully changed the structure of activated carbon by controlling some process conditions to obtain activated carbon with high surface area, porous structure, and high adsorption volume [6]. However, it can be seen that when implementing the physical activation method, the production conditions such as long time, high carbonization and activation temperatures are difficult and energy-consuming [16,18].

This article presents a study on the production of activated carbon from PET waste using physical activation method with CO_2 gas to find the optimal time and temperature. At the same time, it presents some results of the application of the obtained activated carbon in the adsorption and treatment of methylene blue dye in water.

2. Experimental section

2.1 Materials

Waste PET plastic is collected in the form of plastic bottles. The plastic bottles are then cut into small pieces of 1-3 mm, washed and dried. Crystalline iodine, $Na_2S_2O_3 \cdot 5H_2O$, starch, CO_2 gas, and methylene blue were purchased from Xilong Company, China.

2.2 Activated carbon fabrication

Carbonization: Weigh the appropriate amount of PET plastic and put it into a ceramic boat, then place the ceramic boat into the tube furnace. Throughout the heating process, the environment inside the tube furnace is a continuously blowing CO_2 gas environment. The carbonization phase is carried out within a certain period of time and at a specific temperature. After the heating process is complete, the resulting carbonized product is cooled down [7,15].

Yield of carbonization process:

$$H = \frac{m_{carbon}}{m_{plastic}} \times 100\%$$

Carbon activating process: The activated carbon product is baked in a tube furnace (with a CO_2 gas environment) for a specified time and temperature [7,15]. After the activation time, the product is cooled. Then, the activated carbon is further crushed into a fine powder. The activated carbon samples in the form of a fine powder are tested for their iodine number, and the optimized sample is selected for structural analysis and surface area measurement.

Yield of activating process:

$$H = \frac{m_{activated\ carbon}}{m_{carbon}} \times 100\%$$

The physical activation process is carried out by using CO_2 gas, which will act as an activating gas. CO_2 will react with the carbon in the material to create pores that increase the specific surface area and also prevent the infiltration of oxygen gas (O_2) [16].

$$C + CO_2 \longrightarrow 2CO \qquad \Delta H = +159 \text{ kJ/mol}$$

2.3 Materials characterizations

The morphological surface structure characteristics of the activated carbon were observed using a scanning electron microscope (SEM) from HITACHI S-4800 (Japan). The functional groups on the surface of the activated carbon were investigated using Fourier-transform infrared spectroscopy (FTIR) from Perkin Elmer, model Spectrum Two (UK). X-ray diffraction (XRD) measurements were carried out using a machine from X'Pert PRO Panalytical PW3040/60 (Netherlands) with Cu-Kα 0.15405 nm radiation source to study the crystallinity of the samples. The physisorption-desorption method of nitrogen was used with a Tristar 3000-Micromeritics machine to determine the BET surface area of the activated carbon samples.

2.4 Methylene blue (MB) adsorption behaviour of prepared activated carbon

Influence of solution pH: 10 mg of activated carbon was added to 10 mL of 10 ppm MB solution with varying pH values, and the adsorption time was 20 minutes.

Influence of activated carbon dosage: 10 mg, 20 mg, 30 mg, 40 mg, and 50 mg of activated carbon were added to 10 mL of 10 ppm MB solution at pH 7, and the adsorption time was 20 minutes.

Influence of adsorption time: 10 mg of activated carbon was added to 10 mL of 10 ppm MB solution at pH 7 for different periods of time: 15 minutes, 20 minutes, 25 minutes, 30 minutes, 35 minutes, and 40 minutes.

Influence of MB solution concentration: 10 mg of activated carbon was added to MB solution with varying concentrations at pH 7 for 35 minutes.

The adsorption samples were stirred on a stirrer. After stirring, the solid was filtered out, and the resulting solution was measured for absorbance using a UV-VIS machine.

From these results, the adsorption efficiency of the activated carbon was calculated using the formula:

$$H = \frac{C_o - C}{C_o} \times 100\%$$

where: Co, C are the initial and after-adsorption concentrations of MB (ppm); H: Yield (%).

3. Results and discussion

The effect of temperature and time on the carbonization stage

Table 1 is the results of the effect of temperature and time on the carbonization stage. The investigation of the heating temperature from 400 °C to 600 °C for a duration of 15 minutes shows a gradual decrease in efficiency, and a sudden drop in efficiency is observed at 600 °C. This can be explained by the fact that before 600 °C, volatile substances and water vapor were easily released from the material, and at 600 °C, the formation of pores began to develop the porous structure of the activated carbon, resulting in a significant change in mass. Therefore, the temperature of 550 °C was chosen as the temperature for the carbonization process.

Table 1. Effect of temperature on the carbonization yield.

Samples	Temperature (^{o}C)	Time (minutes)	yield (%)
4015	400	15	13.98
4515	450	15	13.67
5015	500	15	13.69
5515	550	15	13.45
6015	600	15	12.5

The effect of carbonization time on the conversion yield was also studied and the result is shown in **Table 2**. A study of the carbonization time at 550 °C from 15 to 30 minutes showed that as the time increased, the carbonization efficiency decreased gradually. This can be explained by the fact that as the time increases, the decomposition of the structure and the volatilization of the substances become

greater. However, at 550 °C, the efficiency of the process did not change significantly, so the choice of the carbonization time period is 15 minutes.

Table 2. Effect of carbonization time on the conversion yield.

Samples	Temperature (^{0}C)	Time (minutes)	Yield (%)
5515	550	15	13.45
5520	550	20	13.28
5525	550	25	13.1
5530	550	30	12.97

The effect of temperature and time on the activating stage

From **Table 3** and **Figure 1**, it can be seen that from the temperature range of 600 °C to 800 °C, both the efficiency and iodine number do not change significantly. However, when the temperature reaches 850 °C, the iodine number increases significantly. This can be explained by the fact that at 850 °C, the pore structure is completed and the ability to adsorb iodine reaches the highest efficiency. At 900 °C, the efficiency and iodine number decrease abruptly, indicating that the process has entered the carbonization stage, and the pore structure is destroyed, directly affecting the quality of the resulting activated carbon. Therefore, 850 °C is chosen as the suitable temperature for the activation process.

Based on the results obtained in **Table 4** and **Figure 2**, it can be seen that there is no significant change in performance during the 25-minute activation time compared to the 20-minute time, and the iodine number reaches its highest value. The iodine number and performance decrease at 30 minutes, indicating that the formed pore system has been carbonized, resulting in a decrease in specific surface area. Therefore, 25 minutes is the appropriate activation time.

Table 3. Effect of temperature on the activating stage.

Samples	Carbonization temperature (^{0}C)/ time (minutes)	Activating temperature (^{0}C)	Activating time (minutes)	Yield (%)
AC6020	550/15	600	20	11.31
AC6520	550/15	650	20	11.30
AC7020	550/15	700	20	11.21
AC7520	550/15	750	20	9.79
AC8020	550/15	800	20	9.5
AC8520	550/15	850	20	8.69
AC9020	550/15	900	20	8.09

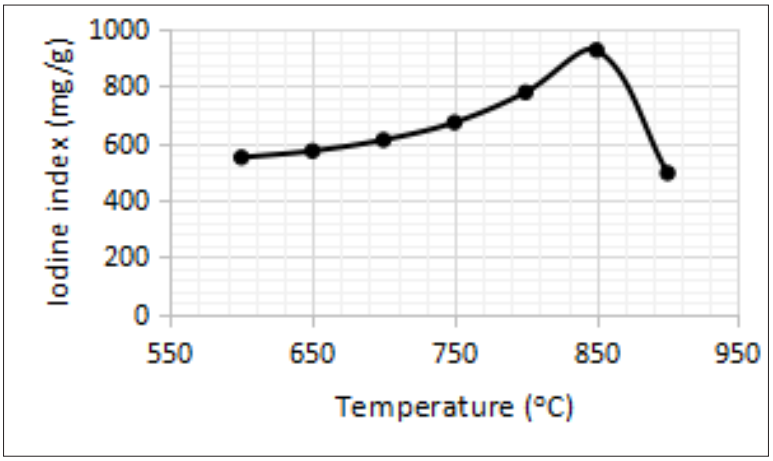


Figure 1. Effect of the activating temperature on the iodine adsorption.

Table 4. Effect of time on the activating stage.

Sample	Temperature (°C)	Time (minutes)	Yield (%)	Iodine index (mg/g)
AC8520	850	20	8.69	927.1
AC8525	850	25	8.57	1003.3
AC8535	850	30	8.02	749.3

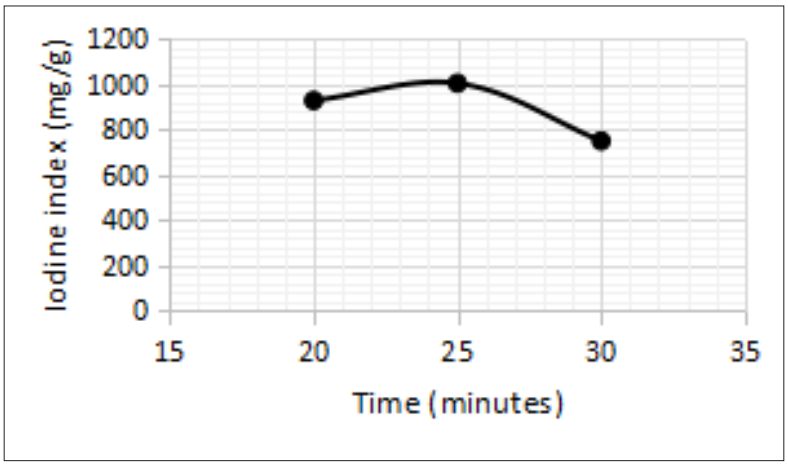


Figure 2. Effect of the activating temperature on the iodine adsorption.

3.1 Characterizations of the activated carbon fabricated using optimized conditions

From **Figure 3**, it can be seen that the X-ray diffraction pattern of the prepared activated carbon sample. The increase in peak intensity in the range of 10-30 degrees is due to the presence of continuous pore voids that scatter X-ray radiation [18]. Subsequently, the XRD peaks weaken, indicating that the activated carbon is in the form of graphite and has many defects on the surface, resulting in a decrease in graphitic crystallization of the activated carbon (JCPSD no. 00-056-0160). Therefore, in the activation process, it is desirable to create more defects on the surface of the carbon to increase the surface area of the activated carbon.

The surface chemical characteristics of the activated carbon are demonstrated through the FTIR spectrum as shown in **Figure 4**. A series of characteristic peaks shown in the graph indicate the presence of various chemical functional groups. The peak at 3447.77 cm^{-1} indicates the -OH group on the surface structure of the activated carbon [14]. The peaks at around 2923.56 and 2871.39 cm^{-1} indicate the appearance of C-H bonds in the benzene and alkane rings. The peak at around 1600 cm^{-1} is characteristic of C-C bonds, while the peak at around 1300 cm^{-1} is characteristic of C-O bonds. The presence of -OH groups may be due to the adsorption of moisture on the activated carbon surface under normal storage conditions, while the presence of C-O groups is likely due to the reaction between C and CO_2, creating a porous structure with the appearance of C-O bonds. The results indicate that the activated carbon obtained is suitable for adsorbing organic compounds, including organic dyes.

The surface morphology of the activated carbon made from PET waste plastic is shown in the SEM image (**Figure 5**). It can be seen that the surface of the activated carbon is relatively smooth, with no clear presence of pores, voids, or channels on the surface.

The surface area is one of the important factors directly affecting the adsorption capacity of activated carbon. The principle is the equal heat of adsorption of nitrogen used to determine the volume of a monolayer. Knowing the covering area of N_2 in the adsorbed state, the specific surface area of the adsorbent can be calculated [19]. **Figure 6** shows the nitro-

gen adsorption isotherm of activated carbon derived from PET waste was measured at 77.3 K. The BET surface area measurement results showed that the activated carbon sample carbonized at 550 °C for 15 minutes and activated at 850 °C for 25 minutes had a specific surface area of 703.4 m^2/g.

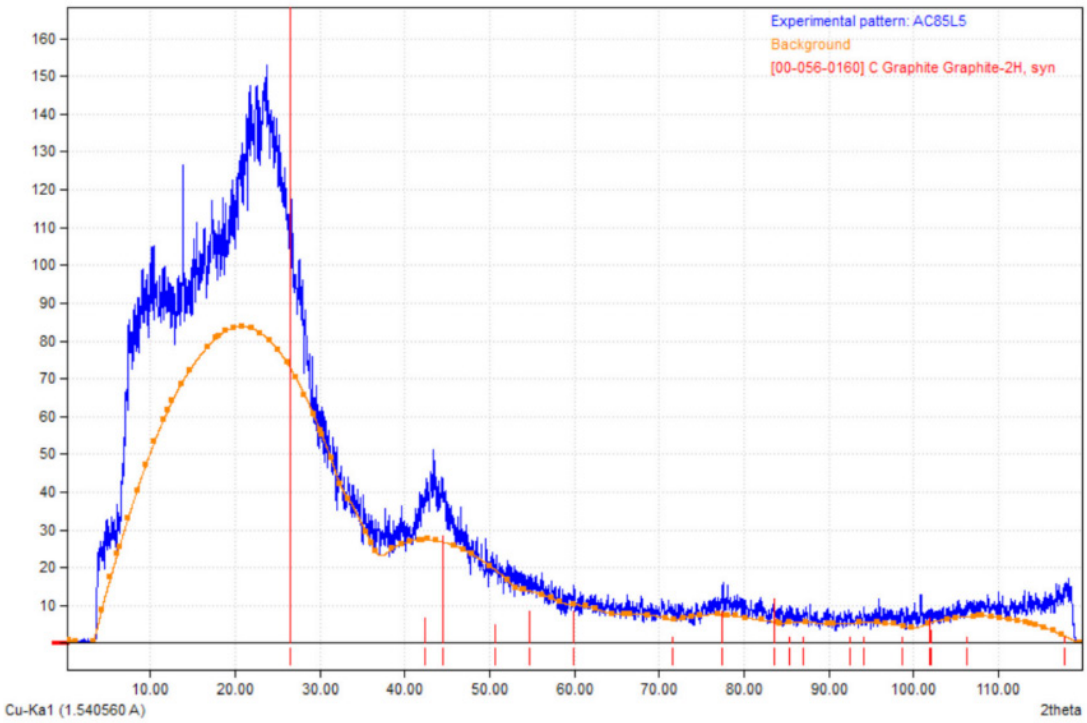
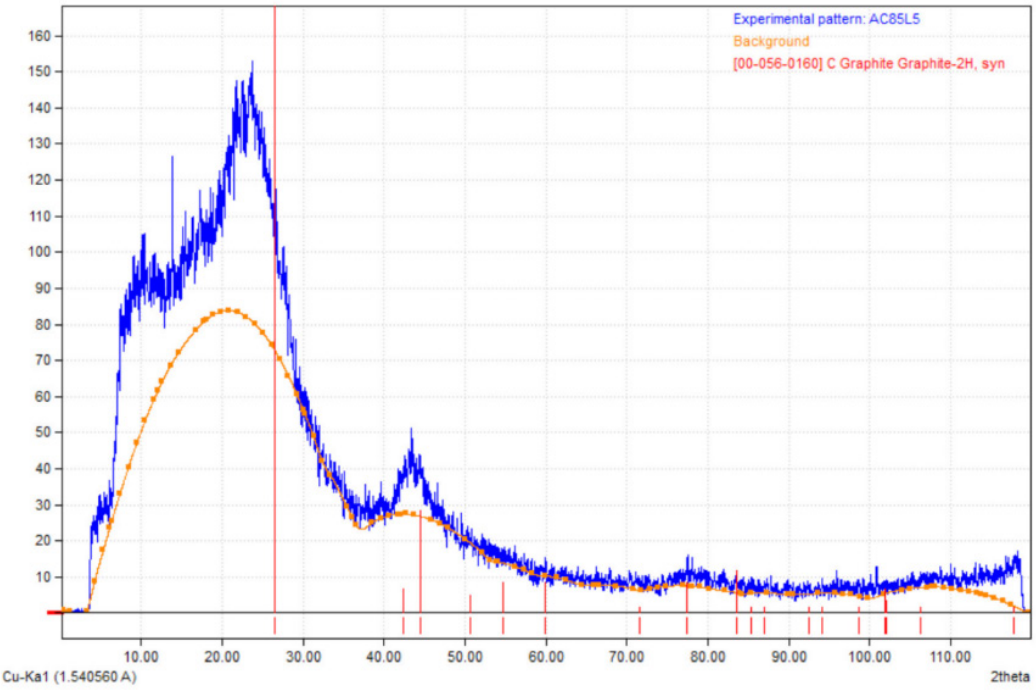


Figure 3. XRD pattern of the activated carbon fabricated from PET plastic using physical activation route at temperature 850 °C for 20 minutes.



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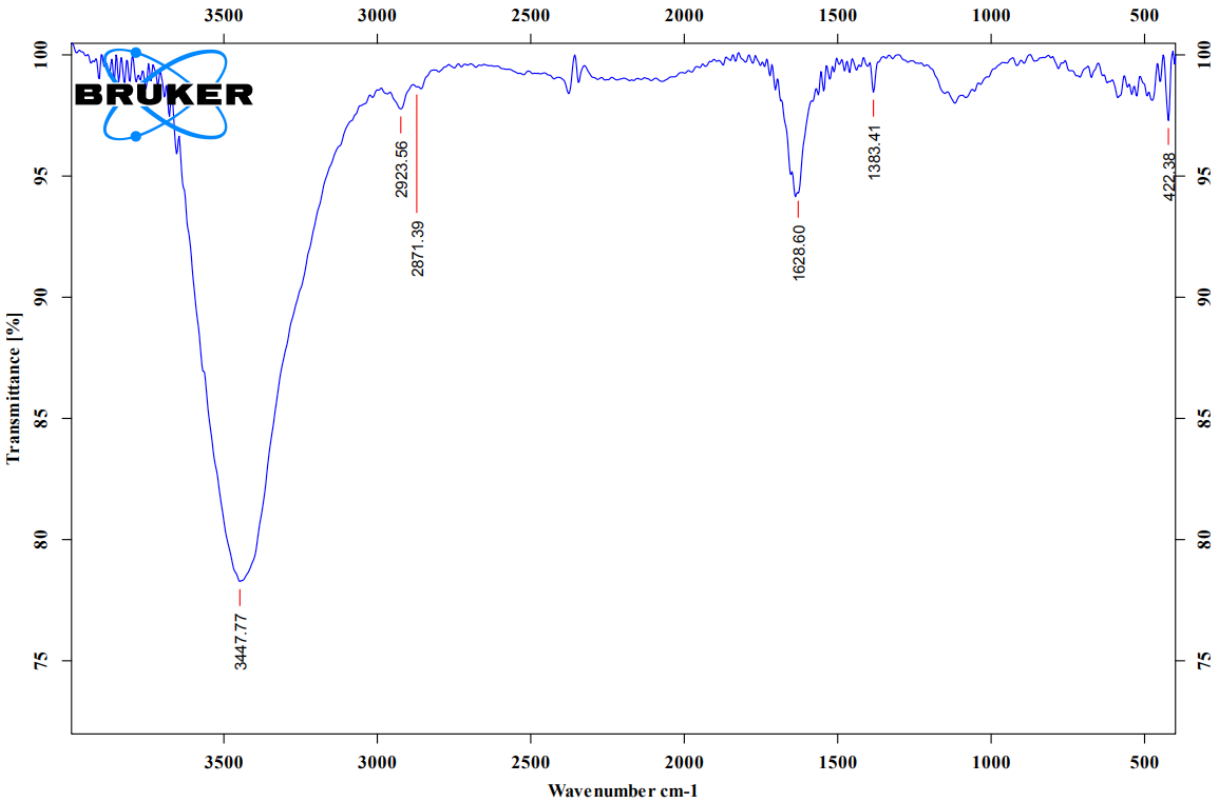

Figure 4. FTIR spectrum of the activated carbon fabricated from PET plastic using physical activation route at temperature 850 °C for 20 minutes.

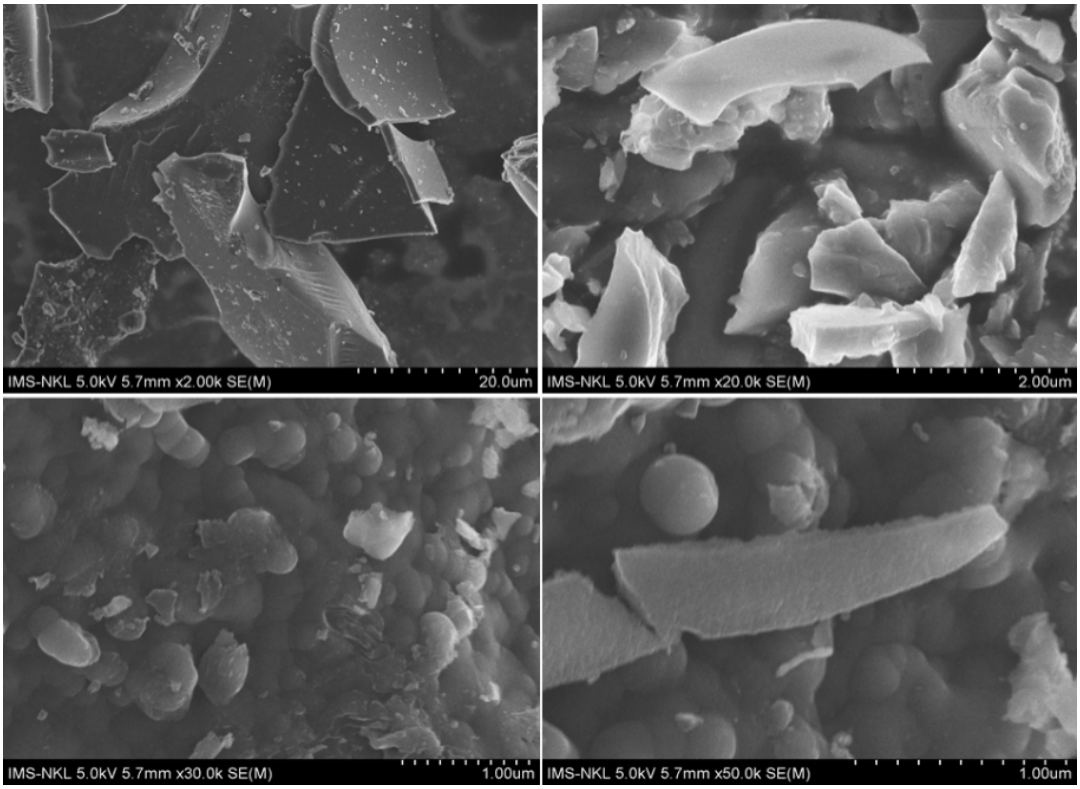

Figure 5. SEM images of the activated carbon fabricated from PET plastic using physical activation route at temperature 850 °C for 20 minutes.

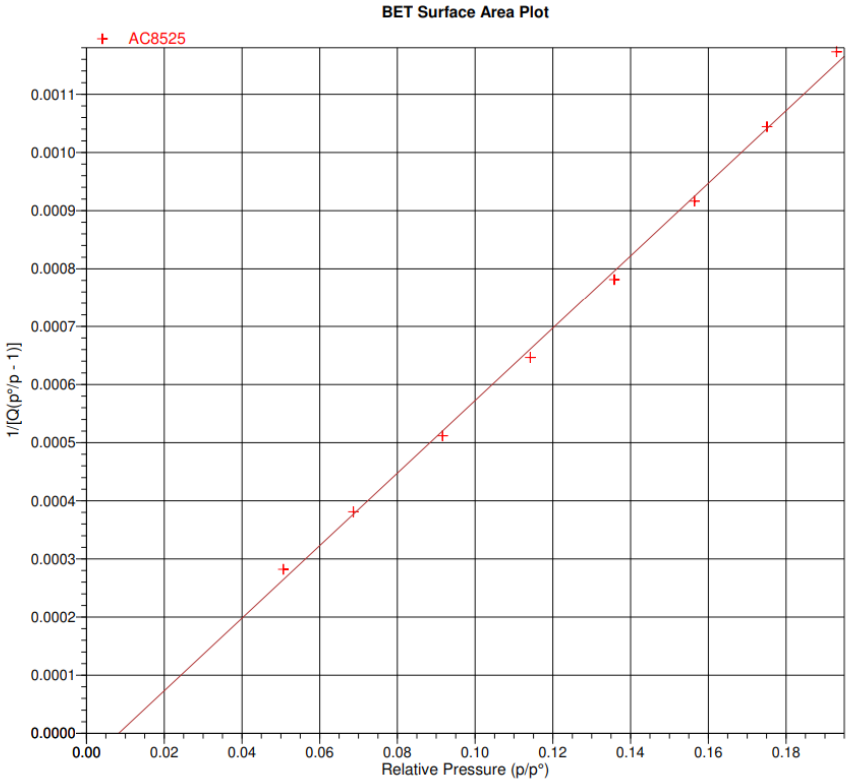

Figure 6. The nitrogen adsorption isotherm of activated carbon derived from PET waste was measured at 77.3 K.

3.2 MB adsorption behaviour of prepared activated carbon

Effect of pH on the MB adsorption efficiency

From **Figure 7**, it can be seen that when pH < 7 (acidic environment), the MB adsorption efficiency of the activated carbon is only below 50%, but when pH ≥ 7, the adsorption efficiency reaches over 80%. The investigation shows that the activated carbon adsorbs well in a pH ≥ 7 environment. Therefore, the next processes will be carried out in a neutral environment, which is suitable for practical conditions in dyeing wastewater treatment.

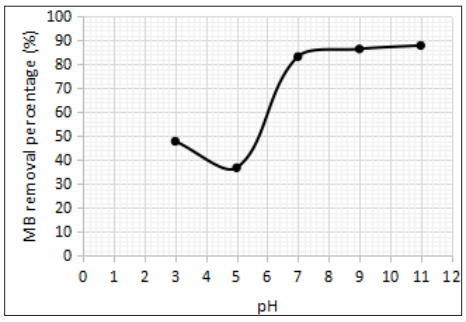

Figure 7. Effect of pH on the MB adsorption efficiency.

Effect of amount of activated carbon on the MB adsorption efficiency

From the survey results, we can see that the higher the mass of activated carbon, the higher the adsorption efficiency. **Figure 8** reveals the effect of the amount of activated carbon on the MB adsorption efficiency. Specifically, with a mass of 0.01 g of carbon, the adsorption efficiency is only 83.212%, while increasing the mass of carbon to 0.02 g results in an efficiency of 94% and there is not much change when increasing the mass of carbon further. Therefore, a mass of 0.02 g of carbon is a suitable condition for weighing and investigating further adsorption processes.

Effect of time on the MB adsorption efficiency

The effect of time on the MB adsorption efficiency was also studied as shown in **Figure 9**. From the obtained results, we can see that the adsorption efficiency is proportional to the adsorption time. The longer the adsorption time, the higher the efficiency and it gradually stabilizes. Specifically, during the adsorption time from 15 to 35 minutes, the efficiency

gradually increases from 87.115% to 95.033%, and at 40 minutes, the adsorption efficiency stabilizes at 95.033%. Therefore, at the time of 35 minutes, the adsorption process reaches equilibrium.

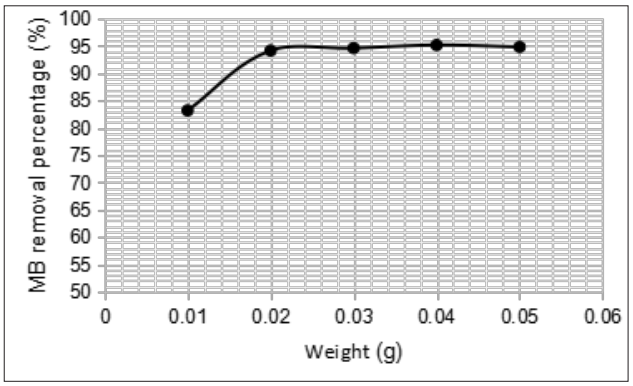

Figure 8. Effect of amount of activated carbon on the MB adsorption efficiency.

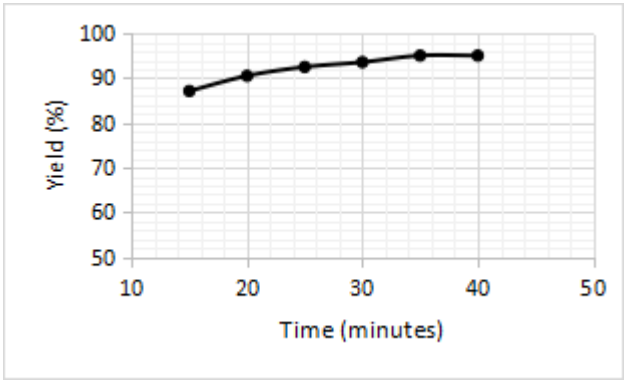

Figure 9. Effect of time on the MB adsorption efficiency.

Effect of MB concentration on the MB adsorption efficiency

Illustrated in **Figure 10** is the effect of MB concentration on the MB adsorption efficiency. The experimental results show that under the same conditions, the adsorption capacity decreases as the MB concentration increases. When increasing the concentration of the solution from 10 ppm to 20 ppm, the efficiency decreases insignificantly (about 4%). However, when the concentration reaches 30%, the color adsorption efficiency drops to 81.34%, and when the concentration reaches 40%, the adsorption efficiency is only 61.18%.

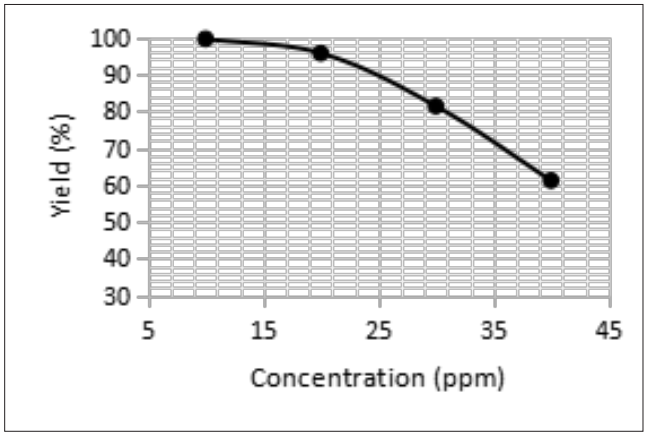

Figure 10. Effect of MB concentration on the MB adsorption efficiency.

4. Conclusions

The successful synthesis of activated carbon materials from PET waste using a physical activation method provides an eco-friendly solution to manage PET waste while producing valuable material for various applications. The specific surface area of 703.4 m^2/g suggests that the activated carbon has a high adsorption capacity for various pollutants. The good adsorption capacity of the PET waste-derived activated carbon for MB in a pH $\geq$ 7 environment indicates its potential as an adsorbent for wastewater treatment. The Langmuir isotherm model provides a good fit for the MB adsorption process, and the maximum adsorption capacity of 18.284 mg/g suggests that the PET waste-derived activated carbon is an efficient adsorbent for MB removal. Overall, this study demonstrates the potential of PET waste-derived activated carbon as a sustainable and effective adsorbent for environmental applications.

Author Contribution

Thu Hanh Pham Thi is responsible for all works that have been done for this work, including conceptualization, experiments, analysis, characterization, manuscript preparation, and submission.

Conflict of Interest

There is no conflict of interest.

Acknowledgement

This work was financially funded by The Ministry of Natural Resources and Environment (No. TNMT.2022.05.04).

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