Activated carbon materials-derived from polyethylene terephthalate (PET) plastic waste using H_3PO_4 activation for Rhodamine B removal in aqueous solution

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ABSTRACT

Plastic items, which offer convenience, are ubiquitous in several manufacturing sectors and in everyday life. Polyethylene terephthalate (PET) is a highly popular synthetic plastic that is seeing a growing demand. Annually, a substantial quantity of PET plastic garbage is released into the environment. Hence, it is imperative to devise an efficient remedy for the disposal of PET plastic waste. This work employed PET waste plastic to produce activated carbon by the chemical activation method. The activating agent utilized was H_3PO_4 acid. An investigation was conducted to determine the impact of the impregnation rate of PET waste plastic with H_3PO_4, as well as the activating temperature and activating time, on the surface areas of activated carbon. The activated carbon was thoroughly analyzed using scanning electron microscopy (SEM), X-ray diffraction (XRD), Fourier-transform infrared spectroscopy (FTIR), and Brunauer-Emmett-Teller (BET) analysis. The resulting product has a porous structure, a developed pore system, and a specific surface area of 892 m^2/g, with effective adsorption capacity for RhB solutions with concentrations below 80 ppm (efficiency above 90%) in a neutral environment according to the Langmuir adsorption isothermal model, with a maximum adsorption capacity of 45.45 mg/g.

Keywords: Plastic waste; Activated carbon; Environmental treatment; RhB removal; Chemical activation.

1. INTRODUCTION

Plastic has become an essential part of life and production. However, plastic is a type of waste that decomposes very slowly. Large pieces of plastic waste are fragmented under mechanical action into small plastic particles less than 5 mm in size and it takes hundreds of years, even thousands of years for a piece of plastic trash to break down in natural conditions. The impact of plastic waste on the environment is huge: water pollution, soil, climate, direct impact on human health and natural landscape. Plastic landfills in particular and waste recycling facilities in general are gradually becoming overloaded. Waste of polyethylene terephthalate (PET) plastic accounts for large amounts of total plastic waste [1-4]. In the face of this problem, there is a need for a better and more effective solution to deal with.

Plastic waste treatment is being actively carried out in many countries around the world, with methods that can be used including recycling, burial, incineration, microbiological degradation and conversion into useful materials. According to statistics, in Europe, about 38% of plastic waste is disposed of, 26% is recycled, and the remaining 36% is for energy recovery [5, 6]. In Vietnam, plastic recycling is slow and inefficient due to difficulties in sorting waste at source. Moreover, plastic after recycling no longer retains its qualities under the effects of heat, so it cannot be reused for enveloping purposes, especially in food packaging or in medical. Therefore, the exploitation of this enormous waste source has and is attracting the greatest interest of regulators. Analysis of carbon content in plastics found that synthetic plastics contain more than 60% carbon [7-10], a lot of scientists around the world have recently focused on developing technology to regenerate plastic waste into carbon-based materials such as graphene [11], carbon nanotubes [12] and especially activated carbon [13]. The most commonly used technology to regenerate plastic waste into activated carbon is chemical activation with acid agent H_3PO_4 [14-16].

This paper presents the study of the production of activated carbon from PET waste plastics by chemical activation with H_3PO_4 acid, as well as some of the results of the application of activated carbon obtained in the adsorption, color processing of Rhodamine B (RhB) dye in water.

2. EXPERIMENTAL SECTION

2.1. Materials

The PET waste that's collected is plastic bottle shells. Then, the plastic bottles are cut in pieces from one to three millimeters, washed and dried. Chemicals: H_3PO_4 85% acid, crystalline iodine 99.9%, $Na_2S_2O_3.5H_2O$ 99.5%, CO_2 99% gas, NaOH 99.5%, Rhodamine B 99 were purchased from Xilong Chemicals (China).

2.2. Fabrication of activated carbon from PET plastic waste

Post-consumer PET plastic waste treated by soaking with H_3PO_4 at a PET:H_3PO_4 weight ratio of 1:1 was calcined in a tube furnace (CO_2 gas environment) at a temperature of 900 °C for 10 minutes. The resulting product, after calcination, was washed with a 5% NaOH solution and distilled water, rinsed several times until the rinsing water reached a neutral pH, and then stopped. After rinsing, the sample was placed in a drying oven at 105 °C for 2 hours. The dried activated carbon was finely ground. The sample was employed for characteristic structural analysis and surface area measurement.

2.3. Characterization of activated carbon

The surface morphology and structure 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 synthesized activated carbon were investigated using Fourier-transform infrared spectroscopy (FTIR) from Perkin Elmer, model Spectrum Two (UK). X-ray diffraction (XRD) measurements were conducted using a machine from X'Pert PRO Panalytical PW3040/60 (Netherlands) with Cu-Kα radiation source at 0.15405 nm to study the crystallinity of the samples. The adsorption-desorption method of nitrogen gas using a Tristar 3000-Micromeritics apparatus was employed to determine the specific surface area (BET) of the activated carbon samples.

2.4. Rhodamine B (RhB) adsorption study

The influence of solution pH: 20 mg of activated carbon was adsorbed in 10 ml of 20 ppm RhB solution at different pH levels, with an adsorption time of 1 minute.

The influence of adsorption time: 20 mg of activated carbon was adsorbed in 10 ml of 40 ppm RhB solution (pH = 7) for different durations: 5 minutes, 10 minutes, 15 minutes, 17 minutes, 20 minutes.

The influence of RhB solution concentration: 20 mg of activated carbon was adsorbed in 10 ml of RhB solution (pH = 7) with varying concentrations for 17 minutes (at room temperature).

Adsorption experiments were conducted using a magnetic stirrer. After stirring, the solid phase was filtered out, and the obtained solution was measured for optical absorption using a UV-VIS spectrophotometer.

From there, the adsorption efficiency of the synthesized activated carbon can be calculated using the formula:

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

Where:

C_o, C are the initial and after adsorption concentrations of RhB (ppm)

H: Removal efficiency (%)

The maximum adsorption capacity of the activated carbon is determined based on the linear

form of the Langmuir isotherm adsorption equation graph:

$$\frac{C_{cb}}{q} = \frac{1}{b.q_{max}} + \frac{1}{q_{max}}.C_{cb} \quad (1)$$

Where:

q, q_{max}: Equilibrium adsorption capacity and maximum adsorption capacity (mg/g);
b: Langmuir constant;
C_{cb}: Concentration of adsorbate at equilibrium (ppm).

3. RESULTS AND DISCUSSION

3.1. Characterizations of activated carbon materials

In this work, activated carbon is synthesized from PET plastic waste with H_3PO_4 as the activating agent (PET waste: H_3PO_4 ratio is 1:1), which is then heated at 900 °C for 10 minutes. Subsequently, it is subjected to analysis of morphology, structure, functional groups present on the surface, and the adsorption study of Rhodamine B dye. The process of producing activated carbon from PET waste has been optimized in previous studies.

The chemical nature on the surface of activated carbon synthesized from PET waste is investigated using FTIR spectroscopy, and the results are depicted in figure 1. In the FTIR spectrum of the synthesized activated carbon sample, a broad peak at 3357 cm-1 indicates the presence of –OH groups on the surface of activated carbon. This presence is attributed to the –OH bonds in the carboxyl group after treatment with H_3PO_4 and from adsorbed water in the structure of activated carbon. Peaks at around 1570 cm-1 are characteristic of C=C bonds. Vibrations at around 1164 cm-1 indicate the presence of phosphorus groups in the structure of activated carbon, such as P=O, P-O-C, P=OOH, P-O-P. These results indicate that the activated carbon synthesized from PET waste includes functional groups suitable for the adsorption of organic compounds, including hazardous dyes. The results are confirmed by scanning electron microscopy (SEM) analysis of the synthesized activated carbon sample [10].

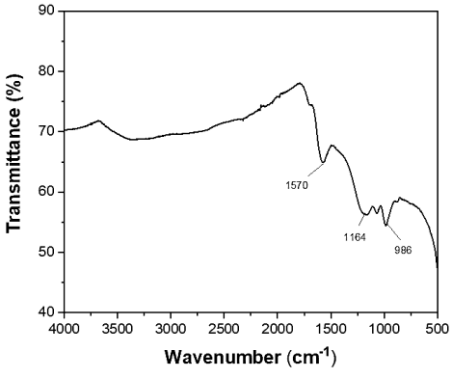


Figure 1. *FTIR spectrum of activated carbon fabricated from PE plastic waste.*

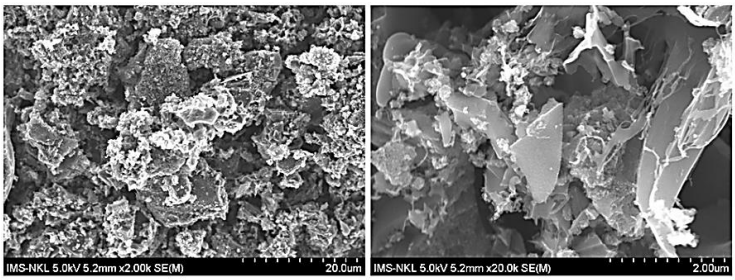


Figure 2. *SEM images of activated carbon fabricated from PE plastic waste.*

The surface morphology of the activated carbon synthesized from PET plastic waste in this study was observed using scanning electron microscopy (SEM). It can be observed that the surface of the activated carbon exhibits numerous pores and voids of various sizes, indicating a large specific surface area and excellent adsorption capacity of the material. The results show the specific surface area of the synthesized activated carbon sample from PET waste.

With the result of the BET surface area measurement, it is found that the activated carbon sample heated at 900 °C for 10 minutes has a specific surface area of 892 m^2/g. The adsorption-desorption method using a Tristar 3000-Micromeritics apparatus was employed to determine the specific surface area (BET) of the activated carbon sample.

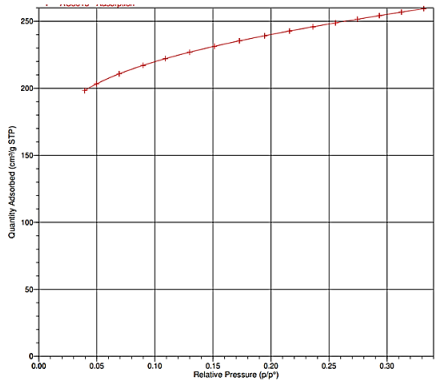


Figure 3. *N_2 adsorption-desorption isotherm of activated carbon fabricated from PE plastic waste.*

3.2. Adsorption study

Five beakers containing 10 ml of RhB solution with a concentration of 20 ppm at different pH values of 3, 5, 7, 9, and 11 were prepared and adsorbed with 20 mg of activated carbon for 1 minute (at room temperature). The results are shown in figure 4. From the obtained results, it is shown that the RhB adsorption efficiency varies as the pH of the RhB solution changes, but the variation is not significant. The adsorption efficiency is consistently above 99%. This result indicates that the activated carbon synthesized from PET waste in this study can be used as an effective adsorbent to remove RhB dye without requiring any cost for pH adjustment. Therefore, in the subsequent surveys of the study, experiments will be conducted under a pH condition of 7.

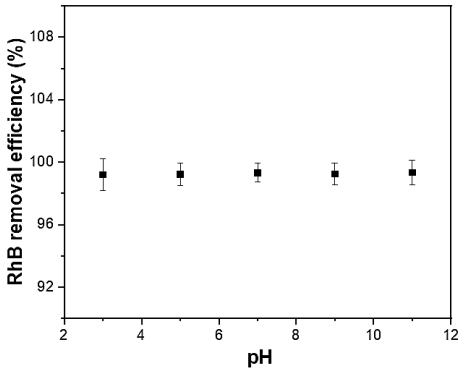


Figure 4. *Effects of solution pH on the adsorption behavior of activated carbon toward RhB.*

Five beakers containing 10 ml of RhB solution with a concentration of 40 ppm at pH = 7 were adsorbed by 20 mg of activated carbon for different durations: 5 minutes, 10 minutes, 15 minutes, 17 minutes, and 20 minutes (at room temperature). The results are shown in figure 5. From the obtained results, it is observed that the adsorption efficiency increases gradually with increasing time. Specifically, within the adsorption time from 5 minutes to 17 minutes, the adsorption

efficiency significantly increases from 98.685% to 99.475%. The adsorption efficiency remains relatively stable from minute 17 to minute 20. The rapid increase in adsorption efficiency in the first 17 minutes is attributed to the rapid adsorption of RhB molecules onto the surface of the activated carbon. Continuing to increase the adsorption time, it is observed that the adsorption pores become filled, and adsorption approaches equilibrium, resulting in little change in adsorption efficiency. Therefore, the adsorption equilibrium time under this experimental condition is determined to be 17 minutes.

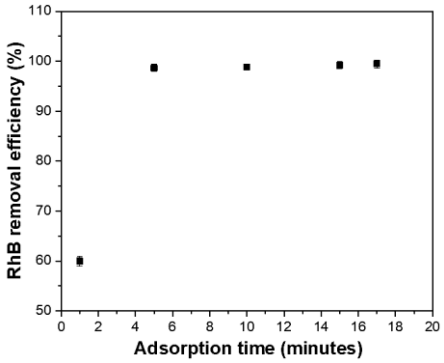


Figure 5. *Effects of adsorption time on the adsorption behavior of activated carbon toward RhB.*

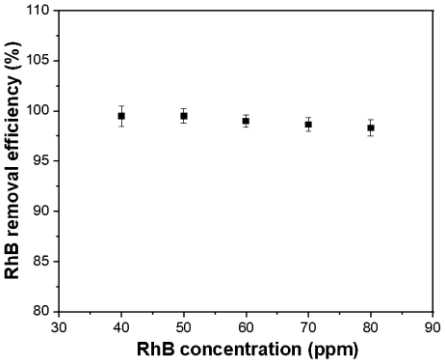


Figure 6. *Effects of RhB concentration on the adsorption behavior of activated carbon toward RhB.*

Five beakers containing 10 ml of RhB solution prepared with different concentrations: 40 ppm, 50 ppm, 60 ppm, 70 ppm, 80 ppm. All solution samples were adjusted to pH 7. Adsorption was carried out using 20 mg of activated carbon for an equilibrium time of 17 minutes (at room temperature). The results are shown in figure 6. The obtained results indicate that the concentration affects the RhB adsorption efficiency of the synthesized activated carbon. As the RhB concentration increases from 40 to 80 ppm, the adsorption efficiency gradually decreases from 99.47% to 98.30%. This can be explained by the occupation of adsorption sites by RhB molecules. During the adsorption process, once the adsorption sites become saturated with RhB, reaching an equilibrium state, they cannot adsorb more RhB molecules. Therefore, as the RhB concentration increases, the adsorption efficiency decreases.

Based on the survey results on the influence of RhB concentration on the adsorption efficiency of activated carbon, we construct the adsorption isotherms according to the Langmuir and Freundlich models. The results are shown in figure 7.

Thus, the Langmuir adsorption isotherm equation for the synthesized activated carbon regarding RhB is:

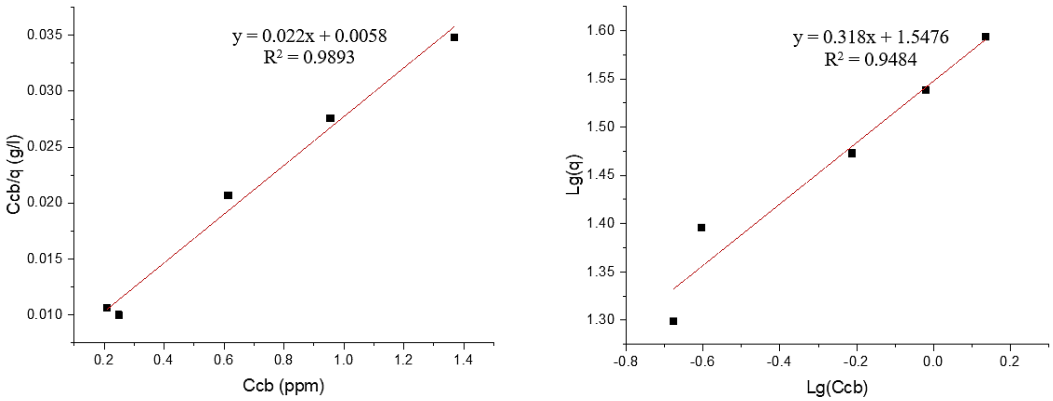
$$y = 0.022x + 0.0058 \text{ with a correlation coefficient } R^2 \text{ of } 0.9893$$

The maximum adsorption capacity (q_{max}) for RhB was determined to be approximately 45.45 (mg/g).

The Freundlich adsorption isotherm equation obtained is:

$$y = 0.318x + 1.5476 \text{ with a correlation coefficient } R^2 \text{ of } 0.9484.$$

From the equation above, we calculate the coefficient $n = 3.14$ ($1 < n < 10$). However, the regression coefficient $R^2 = 0.9484$ is lower than that of the Langmuir adsorption isotherm model (0.9893). Thus, the results obtained from these two models indicate that the Langmuir adsorption isotherm model better describes the RhB adsorption process than the Freundlich model. This suggests that RhB adsorption on the surface of the activated carbon is uniform at all locations on the material surface.



a) Langmuir model. *b) Freundlich model.*

Figure 7. *Adsorption isotherm of activated carbon toward RhB.*

4. CONCLUSIONS

This study successfully synthesized activated carbon from PET plastic waste activated with H_3PO_4 under optimized conditions: activated carbon synthesized from PET waste with H_3PO_4 (PET waste: H_3PO_4 ratio of 1:1) was heated at 900 °C for 10 minutes. The obtained activated carbon has a porous structure, a developed pore system, and a specific surface area of 892 m^2/g, indicating relatively good adsorption capacity. The investigation of Rhodamine B adsorption by the activated carbon demonstrated effective adsorption capability for RhB solutions with concentrations below 80 ppm (efficiency above 90%) in neutral environments. The Rhodamine B adsorption process of the activated carbon synthesized from PET waste was described according to the Langmuir adsorption isotherm model, with a maximum adsorption capacity of 45.45 mg/g. Overall, the findings suggest that activated carbon derived from PET plastic waste can serve as an effective adsorbent for RhB treatment, offering a sustainable solution for wastewater remediation.

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TÓM TẮT

Vật liệu carbon hoạt tính chế tạo từ nhựa thải PET hoạt hóa bằng H_3PO_4 ứng dụng xử lý phẩm màu RhB trong nước

Polyethylene terephthalate (PET) là loại nhựa tổng hợp được sử dụng rất phổ biến hiện nay, với nhu cầu sử dụng ngày càng cao. Hằng năm, một lượng lớn rác nhựa PET được thải trực tiếp ra môi trường. Vì vậy, việc tìm ra một phương pháp xử lý hiệu quả cho việc loại bỏ rác nhựa PET là vô cùng cần thiết. Trong nghiên cứu này, chúng tôi sử dụng rác nhựa PET để sản xuất cacbon hoạt tính bằng phương pháp hoạt hóa hóa học, sử dụng axit H_3PO_4 làm chất hoạt hoá. Khảo sát các yếu tố ảnh bao gồm tỷ lệ ngâm nhựa PET với H_3PO_4, nhiệt độ và thời hoạt hóa đến diện tích bề mặt của cacbon hoạt tính. Tính chất của cacbon hoạt tính được phân tích bằng kính hiển vi điện tử quét (SEM), nhiễu xạ tia X (XRD), phổ hồng ngoại Fourier (FTIR), và phân tích BET (Brunauer-Emmett-Teller). Kết quả cho thấy cacbon hoạt tính được tạo ra từ rác nhựa PET có diện tích bề mặt 892 m^2/g, có khả năng hấp phụ Rhodamine B nồng độ 80 ppm theo mô hình hấp phụ đẳng nhiệt Langmuir nồng độ tối đa 45,45 mg/g.

Từ khóa: Rác thải nhựa; Nhựa PET; Carbon hoạt tính; Hấp phụ môi trường.