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High Porous Activated Carbon from Plastic Waste using Direct Chemical Activation for the Treatment of Organic Dyes in Water

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Abstract: ***Synthetic plastics are ubiquitous across various industries and in everyday life, offering convenience in numerous applications. Among these, polyethylene terephthalate (PET) stands out as one of the most widely used synthetic plastics, with its consumption steadily increasing. However, the substantial volume of PET plastic waste discharged into the environment annually necessitates an effective solution for its management. This study focuses on repurposing PET waste plastic to produce activated carbon using a chemical activation method, employing H_3PO4 acid as the activating agent. The research investigates the influence of factors such as the impregnation rate of PET waste plastic with H_3PO4, activation temperature, and activation time on the surface area of the resulting activated carbon. The characterization of the produced activated carbon was conducted through scanning electron microscopy (SEM), X - ray diffraction (XRD), Fourier - transform infrared spectroscopy (FTIR), and Brunauer - Emmett - Teller (BET) analysis.***

Keywords: Environmental treatment, Activated carbon, plastic wastes, polyethylene terephthalate, organic dyes removal

1. Introduction

Plastic has become an essential part of daily life and manufacturing. However, plastic is a type of waste that decomposes very slowly. Large plastic waste items break down into small plastic particles under mechanical stress, with sizes under 5 mm, and can take hundreds to thousands of years to decompose naturally. The environmental impact of plastic waste is significant, including water, soil, and climate pollution, directly affecting human health and natural landscapes. Plastic landfills and recycling facilities are becoming overloaded. Polyethylene terephthalate (PET) plastic waste accounts for a significant portion of total plastic waste [1 - 4]. Given this issue, there is a pressing need for a better and more effective solution for handling it.

Plastic waste management is actively pursued in many countries worldwide, with methods including recycling, landfilling, incineration, microbial degradation, and conversion into useful materials. Statistics show that in Europe, approximately 38% of plastic waste is landfilled, 26% is recycled, and the remaining 36% is used for energy recovery purposes [5, 6]. In Vietnam, plastic recycling is slow and ineffective due to challenges in waste sorting at the source. Additionally, recycled plastic loses its properties under heat, making it unsuitable for many applications, especially in food packaging or medical fields. Thus, leveraging this massive source of waste has garnered significant attention from policymakers. Analysis of carbon content in various types of plastics reveals that synthetic plastics contain over 60% carbon [7 - 10]. Consequently, many scientists worldwide have focused on researching technologies to convert plastic waste into carbon - based materials such as graphene, carbon nanotubes, and especially activated carbon [12, 13]. A widely used technology for converting plastic waste into activated carbon is chemical activation with H_3PO_4 acid [14 - 16].

This paper presents research on producing activated carbon from PET waste using the chemical activation method with H3PO4 acid. Additionally, it discusses some application results of the obtained activated carbon in adsorption and color removal of methylene blue dye in water.

2. Experimental Section

Materials

PET waste plastic is collected as plastic bottle shells. Then, the plastic bottles are cut into small pieces ranging from 1 to 3mm, cleaned, and dried. H_3PO_4, Iodine, $Na_2S_2O_3.5H_2O$, hồ tinh bột, CO_2, NaOH, and Methylene blue were purchased from Xilong Chemicals (China). All chemicals were utilized as received without any further purification.

Preparation of activated carbon from plastic waste

The PET waste plastic, after being treated by soaking with H3PO4, is heated in a tube furnace (in a CO2 gas environment), with the process carried out at specified time and temperature [14, 16]. The product after heating is washed with a 5% NaOH solution and distilled water, rinsed several times until the rinsing water reaches a neutral pH, then stopped. The sample after rinsing is placed in a drying oven at 105°C for 2 hours. The dried activated carbon is finely ground, and its iodine number is determined. The sample with the best iodine number is selected for characteristic structural analysis and surface area measurement.

Formula for calculating the efficiency of activated carbon: $H = (m_{AC}/m_{plastic}) *100\%$

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 examined by Fourier - transform infrared spectroscopy

(FTIR) using a Perkin Elmer Spectrum Two instrument (UK). X - ray diffraction (XRD) analysis was conducted using an X'Pert PRO Panalytical PW3040/60 machine (Netherlands) with a Cu - Kα radiation source at 0.15405 nm to study the crystallinity of the samples. The physisorption - desorption method of nitrogen, using a Tristar 3000 - Micromeritics instrument, was employed to determine the specific surface area (BET) of the activated carbon sample.

Adsorption study

The influence of the amount of activated carbon: For varying amounts of activated carbon (4mg, 6mg, 8mg, 10mg, 12mg), adsorption was carried out in 10ml of 200ppm MB solution at pH=9 for 7 minutes.

The influence of adsorption time: For 10mg of activated carbon, adsorption was conducted in 10ml of 400ppm MB solution (pH=9) for different durations: 15 minutes, 20 minutes, 25 minutes, 30 minutes, 35 minutes.

The influence of MB solution concentration: For 10mg of activated carbon, adsorption was carried out in 10ml of MB solution (pH=9) with varying concentrations for 25 minutes.

The adsorption samples were agitated using a stirrer. After agitation, the solid part was filtered out, and the resulting solution was subjected to UV - VIS spectrophotometry to measure the absorbance.

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

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

Trong đó:

C_o, C lần lượt là nồng độ ban đầu và sau khi hấp phụ của MB (ppm)

H: hiệu suất (%)

Dung lượng hấp phụ cực đại của cacbon hoạt tính được xác định dựa vào đồ thị của phương trình hấp phụ đẳng nhiệt Langmuir dạng tuyến tính:

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

Trong đó:

q, q_{max}: dung lượng hấp phụ cân bằng và dung lượng hấp phụ cực đại (mg/g)

b: hằng số Langmuir

C_{cb}: nồng độ chất bị hấp phụ ở trạng thái cân bằng (ppm)

3. Results and Discussion

Investigate the factors affecting the synthesis of activated carbon from PET waste activated by H_3PO_4.

Effect of activating temperature

From the survey results, it is observed that the calcination temperature significantly affects the iodine adsorption capacity of activated carbon (Figure 1). Within the temperature range of 400 to 900°C, the iodine number gradually increases and then decreases as the temperature reaches 1000°C. This can be explained by the fact that at temperatures below the optimal range (less than 900°C), the pore structure of the system is not yet fully developed. Based on the synthesis efficiency of the samples, it can be seen that from 400°C to 900°C, the efficiency decreases gradually, with a slight decrease at 800°C and 900°C. However, at 1000°C, the efficiency decreases significantly. Therefore, the specific surface area of the activated carbon continues to improve continuously within the range of 400 – 900°C. However, when the temperature continues to increase to 1000°C, the activated carbon at this point is burned, resulting in ash formation, which leads to the loss of pore structure, causing a significant decrease in efficiency and iodine number. Therefore, the optimal calcination temperature is chosen as 900°C.

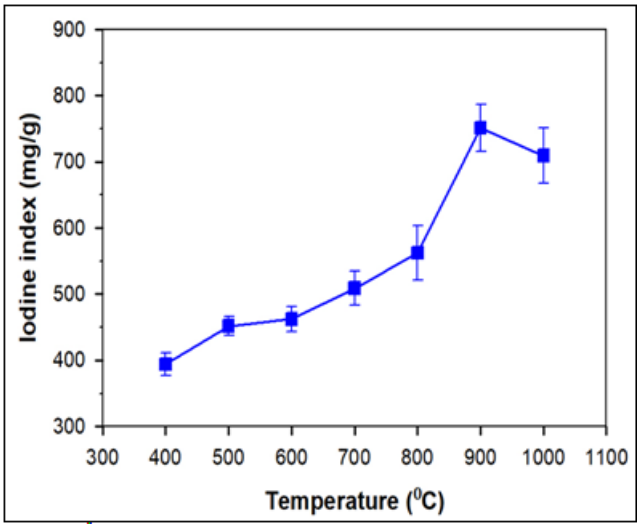

Figure 1: Effect of calcination temperature on the Iodine index of synthesized activated carbon

Effect of PET: H_3PO_4 ratio

From the obtained results, it is noted that the ratio of PET to H_3PO_4 (by weight) affects the iodine adsorption capacity of activated carbon (Figure 2). The iodine number gradually increases until sample IR1 and then decreases. This can be explained by the fact that in sample IR0, the amount of H_3PO_4 is insufficient to induce activation, so when increasing the immersion ratio in sample IR1, the iodine number increases due to better activation. However, when continuing to change the immersion ratio in samples IR2, IR3, IR4, the iodine number decreases because the amount of H_3PO_4 increases, causing more coverage of the plastic particles by H_3PO_4, which agglomerates and hinders the formation and development of pores, thereby reducing activation efficiency and resulting in a gradual decrease in the iodine number. Therefore, the immersion ratio of PET to H_3PO_4 (by weight) is chosen as 1: 1 for activated carbon synthesis.

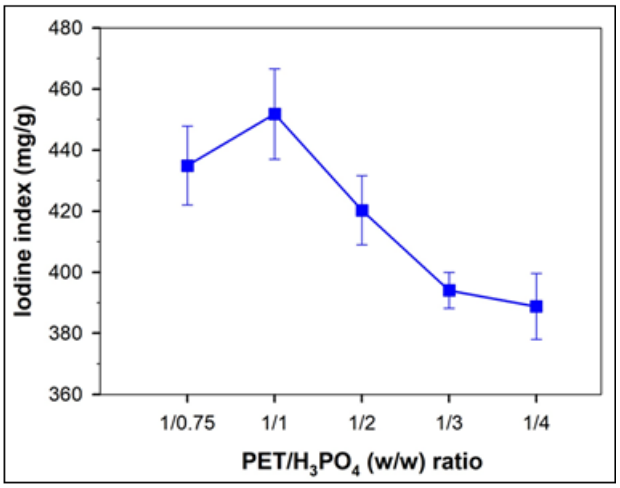

Figure 2: Effect of PET/H_3PO_4 (w/w) ratio on the Iodine index of synthesized activated carbon.

Effect of activating time

From the survey results, it is observed that there is a variation in the iodine number of activated carbon at different calcination times (Figure 3). From 5 to 10 minutes, the iodine number gradually increases, while it decreases from 5 to 25 minutes. Regarding efficiency, there is a gradual decrease as the time increases from 5 to 25 minutes. This variation can be explained by the fact that during activation and synthesis of activated carbon within the first 10 minutes of calcination, the pore structure of the material is not yet fully developed. However, when the calcination time exceeds 10 minutes, carbonization of the activated carbon occurs, resulting in a significant reduction in both efficiency and iodine number. Therefore, the optimal calcination time is chosen as 10 minutes.

Activated carbon synthesized from PET waste with H_3PO_4 as the activating agent (PET: H_3PO_4 ratio of 1: 1) is calcined at 900°C for 10 minutes. Subsequently, it is subjected to analysis of morphology, structure, functional groups present in the charcoal, and examination of adsorption of methylene blue dye.

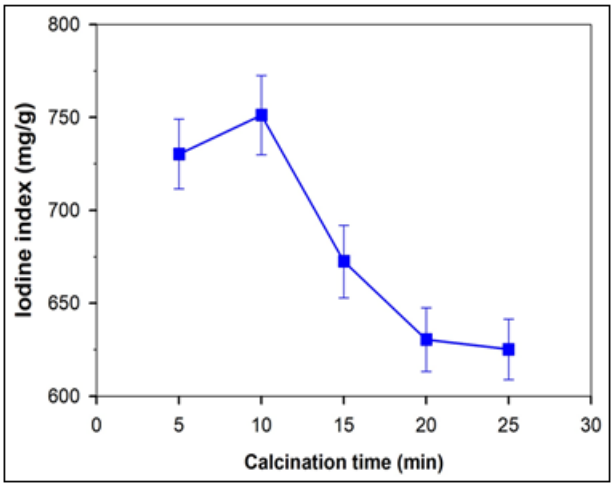

Figure 3: Effect of calcination time on the Iodine index of synthesized activated carbon

Characterization of prepred activated carbon from plastic waste

Figure 4 shows the X - ray diffraction pattern of the synthesized activated carbon sample. The peaks in the X - ray diffraction pattern exhibit increased intensity within the range of 10 - 30 degrees, indicating the presence of continuous pores scattering the X - ray radiation [17]. Subsequently, the X - ray diffraction peaks weaken. It can be observed that the resulting activated carbon has a graphitic structure, suggesting the presence of defects on the graphite walls, leading to a decrease in graphitic crystallinity of the activated carbon (JCPSD no.00 - 056 - 0160). Thus, during the activation process, it is desired to create more defects on the carbon walls to increase the surface area of the activated carbon.

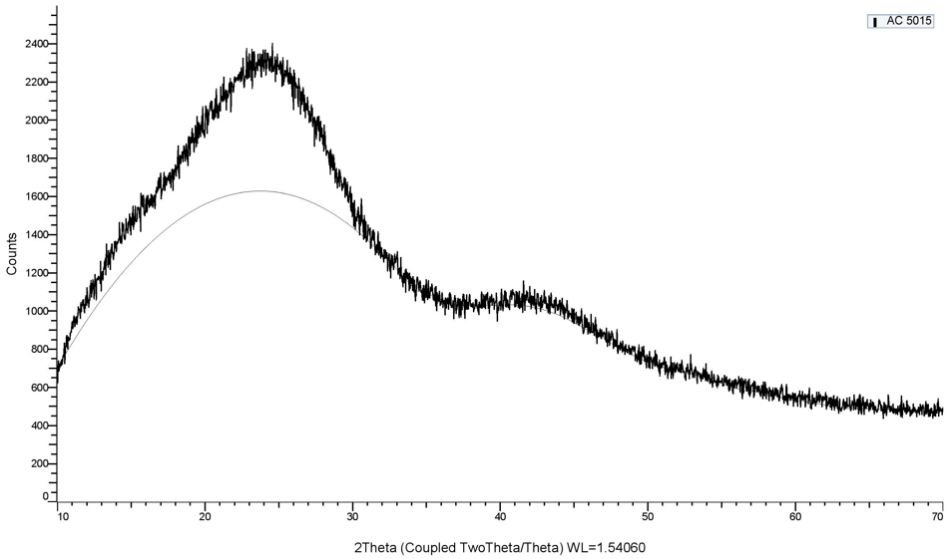

Figure 4: XRD pattern of synthesized activated carbon calcinating at 900 ^{0}C for 10 min.

The chemical nature of the surface on the activated carbon synthesized from PET waste was examined using FTIR spectroscopy, and the results are depicted in Figure 5. In the FTIR spectrum of the synthesized activated carbon sample, a broad peak appears at 3356.56 cm $^{-1}$, indicating the presence of hydroxyl groups (- OH) on the surface of the

activated carbon [18]. This presence is attributed to the -OH bonds in the carboxyl groups after treatment with H3PO4 and from adsorbed water in the structure of the activated carbon. Peaks at around 1569.84 cm $^{-1}$ correspond to characteristic C=C bonds [19]. Vibrations at 1163.61 cm $^{-1}$ indicate the presence of phosphorus groups in the structure of the activated carbon, such as P=O, P - O - C, P=OOH, P - O - P [20]. These results indicate that the activated carbon synthesized from PET waste includes functional

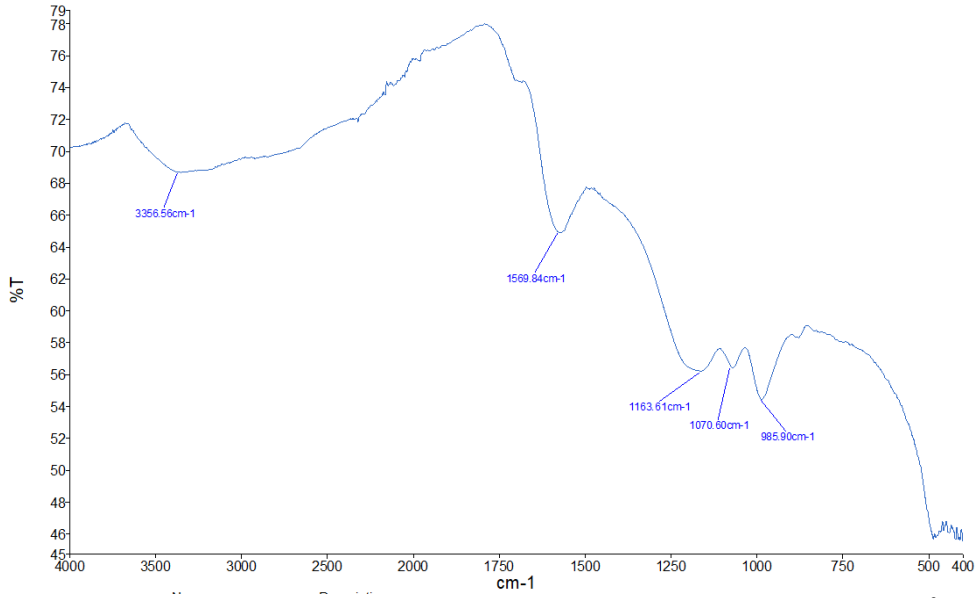

Figure 5: FTIR spectrum of synthesized activated carbon powdered sample calcinated at 900 ^{0}C for 10 min.

The surface morphology of the activated carbon synthesized from PET waste in this study was observed using scanning electron microscopy (Figure 6). It can be observed that the surface of the activated carbon exhibits numerous fissures, grooves, and pores of varying sizes, indicating a large specific surface area and excellent adsorption capability of the material. Surface area is one of the crucial factors directly influencing the adsorption capacity of activated carbon. The principle is based on the equilibrium adsorption of nitrogen to determine the volume of monolayer coverage. Knowing the surface coverage area of N_2 in the adsorbed state allows for the calculation of the specific surface area of the adsorbent material [21]. From the results of the BET surface area measurement, it can be seen that the activated carbon sample synthesized from PET waste impregnated with H_3PO_4 at a ratio of 1: 1, calcined at 900°C for 10 minutes, has a specific surface area of 1700 m^2/g.

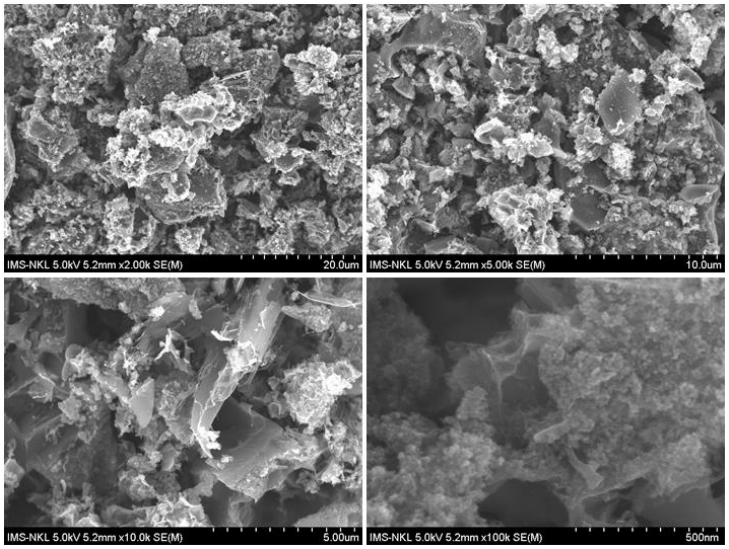

Figure 6: SEM micrographs of synthesized activated carbon sample calcinated at 900 ^{0}C for 10 min.

Adsorption behaviour of prepared activated carbon for MB

From the Figure 7a, it is evident that the mass of activated carbon has an impact on the adsorption efficiency of MB. As the mass of the adsorbent material increases from 2 mg to 12 mg, the adsorption efficiency gradually increases. With an adsorbent mass of 10 mg and 12 mg, the efficiency stabilizes. Based on the obtained results, the appropriate mass of activated carbon chosen for further experiments is 10 mg.

The results from Figure 7b indicate that the adsorption time affects the MB adsorption efficiency of the activated carbon. The longer the contact time between MB and activated carbon, the higher the adsorption efficiency. At the 25 - minute mark, the adsorption efficiency stabilizes as the adsorption process reaches equilibrium. Therefore, the adsorption time chosen for further experiments is 25 minutes.

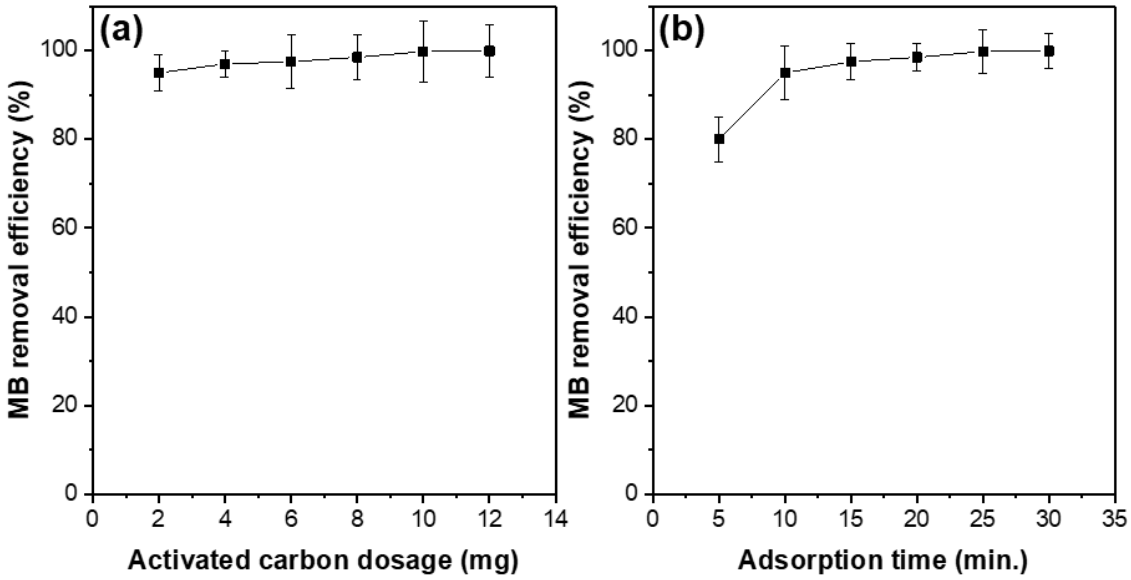

Figure 7: Effect of (a) activated carbon's dosage and (b) adsorption time on the MB removal efficiency

The results obtained indicate that the concentration has an impact on the MB adsorption efficiency of the synthesized activated carbon. When the MB concentration increases from 10 to 400 ppm, the adsorption efficiency gradually decreases from 99.9% to 98%. This decrease occurs because during the adsorption process, at a certain stage, the adsorption sites become saturated with MB, reaching equilibrium. At this point, no further MB adsorption occurs. Therefore, when the MB concentration increases, the adsorption efficiency decreases.

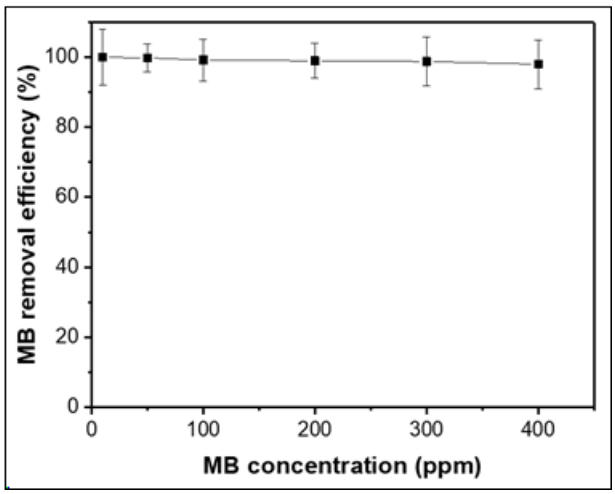

Figure 8: Effect of MB concentraions on the MB removal efficiency

4. Conclusion

In this study, the research team successfully synthesized activated carbon material from PET waste using the chemical activation method with H_3PO_4 under optimal conditions: PET: H_3PO_4 ratio = 1: 1, calcination at 900°C for 10 minutes. The resulting activated carbon has a porous structure with a specific surface area of 1700 m^2/g. The activated carbon from PET waste exhibits excellent adsorption capacity for methylene blue organic dye, with adsorption efficiency reaching nearly 100% after 15 minutes of adsorption. With a simple manufacturing process, activated carbon material synthesized from PET waste not only contributes to reducing the environmental impact of solid waste but also produces valuable products with good environmental remediation capabilities.

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