

Porous activated carbon spheres derived from resorcinol-formaldehyde resin with high performance for methylene blue removal

Do Dinh Trung¹, Vu Dinh Thao², Nguyen Ngoc Linh³, Nguyen Duc Trung⁴,
Nguyen Ngoc Tue^{4,*}

¹*Institute of Tropical Durability, Joint Vietnam - Russia Tropical Science and Technology Research Center, No. 63, Nguyen Van Huyen Street, Nghia Do Ward, Cau Giay District, Ha Noi, Viet Nam*

²*Faculty of Physical and Chemical Engineering, Le Quy Don Technical University, Hoang Quoc Viet, Ha Noi, Viet Nam*

³*Faculty of Pharmacy, Thanh Do University of Education, Kim Chung Ward, Hoai Duc District, Ha Noi, Viet Nam*

⁴*School of Chemistry and Life Sciences, Hanoi University of Science and Technology, Ha Noi, Viet Nam*

*Emails: tue.nguyennhoc@hust.edu.vn

Received: 18 December 2023; Accepted for publication: 5 May 2025

Abstract. Porous carbon materials offer numerous practical uses, particularly in the field of adsorbents due to their remarkable characteristics such as low density, high surface area, strong electrical conductivity, and many other benefits. The findings on synthesizing porous carbon materials using a resorcinol-formaldehyde polymerization reaction, a gelation process, and pore water evaporation at high temperature are presented in this study. The procedure was carried out in an atmosphere containing an inert gas. Analyses of the newly created materials included X-ray diffraction (XRD), scanning electron microscopy (SEM), energy dispersive X-ray spectroscopy (EDX), calculation of specific surface area using the BET equation, and many other methods. A porous carbon material with a specific surface area of 801.92 m²/g can be feasibly manufactured.

Keywords: absorption material, porous carbon, resorcinol, formaldehyde, environment treatment.

Classification numbers: 2.6.1, 2.10.1, 3.4.2

1. INTRODUCTION

The revolution of new materials has effectively solved the problem of adsorption science, with adsorbents such as activated carbon, zeolite, and others being improved to meet the reality of more serious pollution treatment with more important significance [1]. Carbon materials, such as activated carbon, graphene, and carbon nanotubes, are some of the most common types of adsorbents. This is because carbon materials have a high organic matter adsorption capacity, are renewable, and are safe for the environment. The specific surface area, average pore diameter,

and total porosity volume of carbon materials are the primary factors that determine the adsorption effectiveness of carbon materials [1, 2].

Porous carbon with large specific surface area (between 400 and 1000 m²/g) has captured the interest of scientists to a significant degree [3]. It is feasible to produce porous carbon on a large scale from polymer composite materials thanks to its particular new features, and it can also be synthesized from waste plastic sources (foam PET) at a significantly cheaper cost and in a shorter time compared to the production of other materials. Current techniques for creating porous materials not only have the potential to use porous carbon on a large scale, but also help solve the problem of plastic waste, a toxic waste that is destroying our planet. There have been many studies conducted all over the world that have focused on the development of porous carbon materials. The overarching objective of these studies is to create porous carbon materials with increasingly preeminent characteristics and the ability to satisfy environmental remediation needs [4, 5].

There are many studies on the adsorption capacity of methylene blue onto lignin-based porous carbon, activated carbon/cellulose biocomposite films, partially hydrolyzed polyacrylamide/cellulose nanocrystal nanocomposite hydrogels, rice husk, modified sugarcane bagasse [6 - 10]. The results of these studies show that the adsorption capacity of methylene blue onto these materials depends greatly on their surface area, pore size, and many other physicochemical properties.

Because of their exceptional qualities, including their porous structure, high micropore volume, large surface area, controllable pore size distribution, smooth surface, excellent durability, good fluidity, low ash content, and low moisture content, porous activated carbon spheres derived from resorcinol-formaldehyde resin significant are of great significance in the field of adsorption [11 - 14].

2. MATERIALS AND METHODS

This section describes concisely the detail methodology/procedures employed so that anyone wishing to replicate the trial can do so and obtain comparable results. Provide sufficient detail so as to remove any possible ambiguities with respect to design, treatments, measurements, analysis, etc. Where methods employed are commonly known in a given field details should be omitted and the reference given instead. Modifications to known methodology must however be clearly described and explained. One paragraph must not contain only one sentence.

2.1. Materials

The chemicals, instruments and equipment used in this study included: Resorcinol 99.5 % (Xilong, China); formaldehyde 38.5 % (Xilong, China); sodium carbonate 99.2 % (Xilong, China); sodium hydroxide 98.0 % (Xilong, China); methylene blue 99.0 % (Xilong, China); 100 mL beaker (Schott Duran, Germany); volumetric flask (Schott Duran, Germany); 100 mL measuring cylinder (Schott Duran, Germany); crucible (China); glass cuvette (China); WTM inoLab pH 7310 benchtop pH meter (Germany); ultrasonic tank (Elma, Germany); drying cabinet (Ecocell, UK); analytical balance (Sartorius, Germany); heating magnetic stirrer (Hana, China); double distilled water machine (Hamilton, UK); UV-Vis SP60 machine (UK); and GFL 3005 Shaker (Germany).

2.2. Methods

A total of 7.2 grams of resorcinol (R) and 23.3 mL of formaldehyde solution (F) were accurately measured in a molar ratio of 1:5. These substances were combined in a 100 mL beaker. Additionally, 1.0 gram of a 10 % Na₂CO₃ solution was added to the beaker as a reaction catalyst, serving the dual purpose of controlling the size of the primary particles and the density of the RF polymer. Finally, 45.0 grams of double distilled water were introduced into the beaker. The subsequent steps are as follows: The experimental procedure involved activating the magnetic stirrer, gradually increasing the temperature of the reaction block at a rate of 2 °C per minute until reaching 50 °C. Subsequently, the reaction was conducted at a constant temperature of 50 °C for 30 minutes. Stirring was then ceased, and the magnet was removed to allow the solution to stabilize at room temperature for a period of time. Following a 24-hour duration, the resultant composition manifests as a gel in the form of resorcinol-formaldehyde (RF) gel. The stabilized RF gel was subjected to a drying process in a controlled environment at a temperature of 90 °C for 48 hours. Subsequently, the dehydrated RF polymer was obtained by allowing it to drop down to room temperature, resulting in the formation of a porous structure. After that, the RF polymer block was carefully inserted into the porcelain crucible, ensuring that the lid was securely in place. The crucible, together with its contents, was then placed in a furnace under tightly controlled inert gas conditions, namely nitrogen (N₂). The RF polymer underwent pyrolysis, following a prescribed thermal cycle. The heating rate was set at 2 °C per minute, starting at an initial temperature of 40 °C and reaching a final temperature of 300 °C. The furnace temperature was maintained at 300 °C for 150 minutes. Subsequently, the furnace was turned off, allowing the temperature to gradually decline to room temperature over a period of 24 hours. The specimen was extracted from the furnace and submerged in a 100 mL solution of 1 M NaOH for 60 minutes. It was thereafter rinsed and NaOH was eliminated through a series of 4 to 5 washes using double distilled water. The porous carbon material is subjected to a thermal treatment at a temperature of 500 °C for 3 hours. The temperature was incrementally raised at a rate of 2 °C per minute in order to sustain the process of pyrolysis on the sample until it reached a temperature of 700 °C. Subsequently, the temperature was maintained at this level for 3 hours. The furnace was deactivated in order to provide a progressive fall in temperature over a period of 24 hours, resulting in the formation of a porous carbon specimen referred to as CX700. The synthesis diagram of resorcinol-formaldehyde polymer and porous carbon is illustrated in Figure 1.

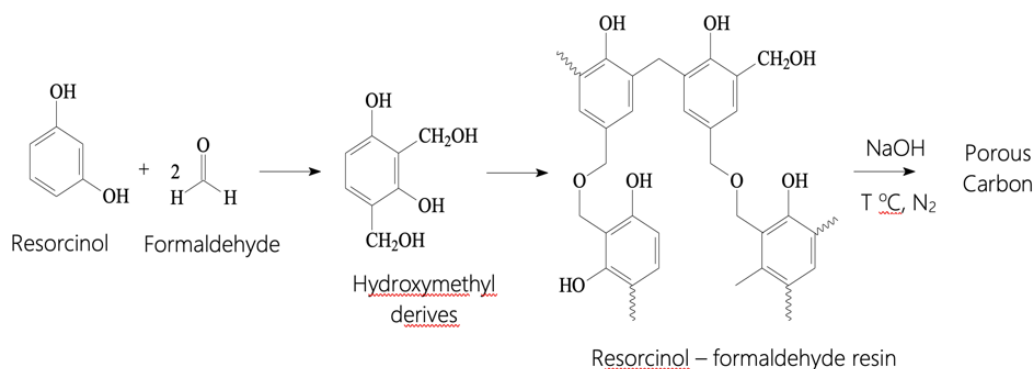


Figure 1. Synthesis diagram of resorcinol-formaldehyde polymer and porous carbon.

2.3. Analytical methods

The specific surface area and pore size of the porous carbon materials were determined by nitrogen isothermal adsorption, the results were calculated according to the BET (Brunauer-

Emmet-Teller) equation. Measurements were performed on a Gemini VII 2390 instrument (Micromeritics Instrument, USA) at the Advanced Institute of Science and Technology, Hanoi University of Technology. Determination of material structure by scanning electron microscope (SEM) images was carried out at the Institute of Materials Science, Vietnam Academy of Science and Technology. The material properties were determined by infrared (IR), energy dispersive X-ray (EDX) and X-ray diffraction (XRD) spectroscopy at the Institute of Chemistry - Materials, Institute of Military Science and Technology.

2.3. Adsorption test

All experiments were carried out under the same optimal conditions, 25 °C and pH 7.0. An amount of 2.0 - 3.0 mg of adsorbent was dispersed into 50 mL of each of the two dyes with various initial concentrations. The volumetric flasks were ultrasonicated for one hour, then were shaken in a gas bath thermostatic oscillator with a shaking speed of 300 rpm until equilibrium was reached. After 1 hour, the adsorption equilibrium was established and residual dye concentration was determined at their specific wavelength (664 nm for Methylene Blue - MB) using a UV-vis spectrophotometer. Concentrations of the dyes were determined using the linear regression equations (obtained by plotting its calibration curve). The amount of the dye adsorbed by the adsorbent (q_e (mg/g)) was calculated by the following formula:

$$q_e = \frac{(C_0 - C_e)V}{W}$$

where C_0 (mg L⁻¹) and C_e (mg L⁻¹) are the initial and equilibrium concentrations of MB, respectively. V is the volume of the solution (L), and W is the weight of the adsorbent (g). Two models, the isotherm models of Freundlich and Langmuir, were used to evaluate the adsorption equilibrium characteristics of MB on the two carbon-based materials. The linear form of the two models are summarized in **Error! Reference source not found.** where q_m (mg/g) is the maximum adsorption capacity and K_L is the Langmuir coefficients. For the Freundlich models, K_F and n are the two corresponding parameters for each studied adsorbent. In addition to giving useful information on reaction pathways, kinetic models were employed to assess the pace of the adsorption process. Langmuir adsorption isotherm is used as following: $\frac{C_e}{q_e} = \frac{1}{q_m K_L} + \frac{C_e}{q_m}$ and Freundlich adsorption isotherm is used as following: $\ln q_e = \ln K_F + \frac{1}{n} \ln C_e$

3. RESULTS AND DISCUSSION

This the morphology structure of the RF and porous carbon samples was observed using SEM and the results are given in Figure 2.

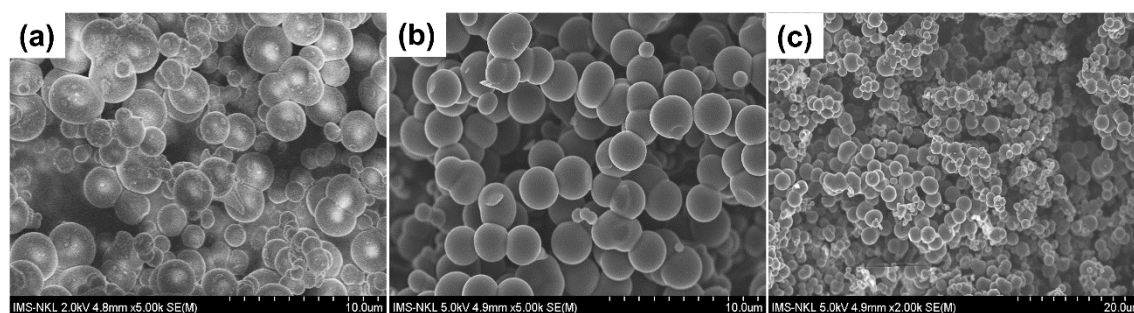


Figure 2. SEM images of materials. (a) RF, (b) porous carbon without using chemical activation agent, (c) porous carbon using NaOH as a chemical activation agent.

It was found that the formed products are spherical in shape (shell-core structure) with diameters ranging from 2 μm to 5 μm , which is similar to the structure reported in certain previously research [15], whereby the particles are stacked on top of one another, and linkages exist at the contact points. The surface of the inactivated carbon samples is fairly flat, with an average size of around 3 μm . For carbon samples activated by NaOH, the shell-core structure is maintained, but the surface is rougher and the average size (about 2.5 μm) is reduced, indicating an oxygen reaction. The chemical exchange between the carbon surface layer and NaOH produces an escape gas that increases the material's porosity and greatly lowers the volume of the carbon sample produced after the reaction. The porous carbon samples formed by using the chemical activator NaOH were used for subsequent studies.

Figure 3 depicts the IR spectrum of resorcinol-formaldehyde (RF). The -OH group is responsible for the broad peak detected in the wavelength band of 3200 - 3400 cm^{-1} due to water adsorption [16]. The peak at 1631.29 cm^{-1} corresponds to the aromatic ring group, whereas the peak at 1473.07 cm^{-1} belongs to the CH_2 group [17]. Many peaks between 950 and 1400 cm^{-1} are attributed to the COC and alkyl-phenyl ether groups. Furthermore, the peak at 780.33 cm^{-1} may represent an out-of-plane vibration group of the OH and CH groups [18]. All the distinctive peaks of the manufactured materials match the previously published IR spectra of RF [19].

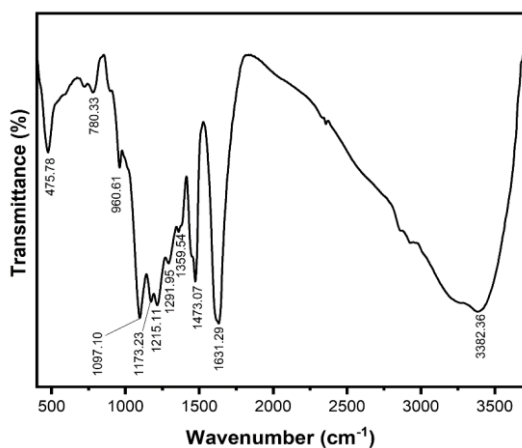


Figure 3. FT-IR spectrum of the RF sample.

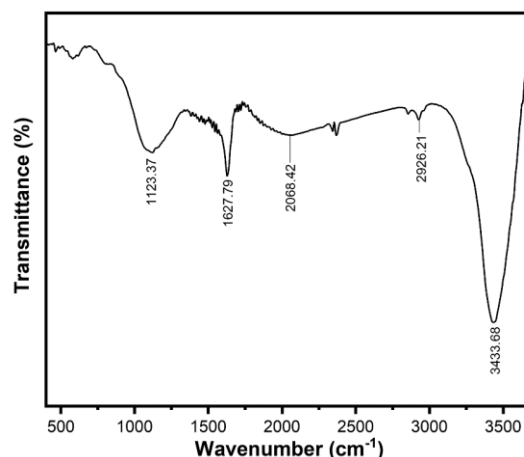


Figure 4. FT-IR spectrum of the porous carbon CX700 sample.

Figure 4 depicts the infrared spectrum of the CX700 sample. Comparing the IR spectra of the resorcinol-formaldehyde polymer to that of the CX700 reveals the presence of stable oscillating functional groups at 1627.79 cm^{-1} and 1123.37 cm^{-1} , respectively. The disappearance of the majority of the other absorption peaks in the CX700 spectrum indicates that the resorcinol-formaldehyde polymer was pyrolyzed at 700 $^{\circ}\text{C}$ and transformed to carbon.

Chemical elements in the CX700 sample were identified by energy dispersive X-ray (EDX) spectroscopy. The results (Figure 5) indicate that the collected sample is mostly constituted of carbon (> 95 %), and with a small amount of oxygen and sodium (10 %), this can serve as a medium for the gelation reaction with activated NaOH and initial Na_2CO_3 .

To further confirm the structure of the synthesized product, X-ray diffraction was taken with a diffraction angle of 2θ from 0 to 70 $^{\circ}$. The results in Figure 6 show that the structure of the resulting porous carbon product is amorphous carbon.

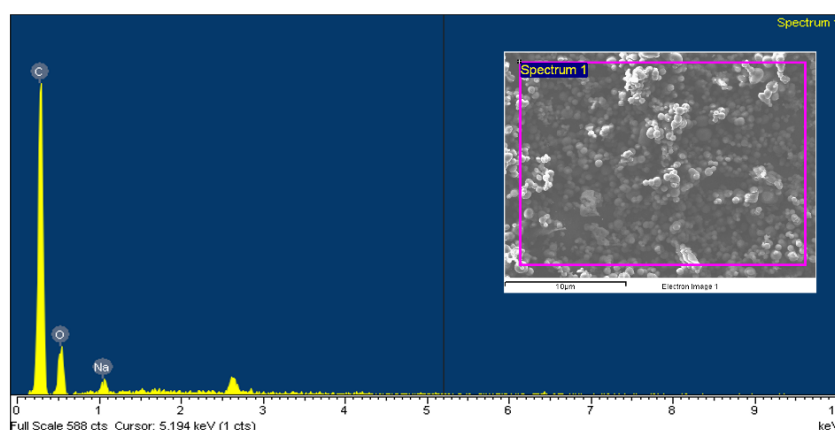


Figure 5. EDX spectrum of the porous carbon CX700.

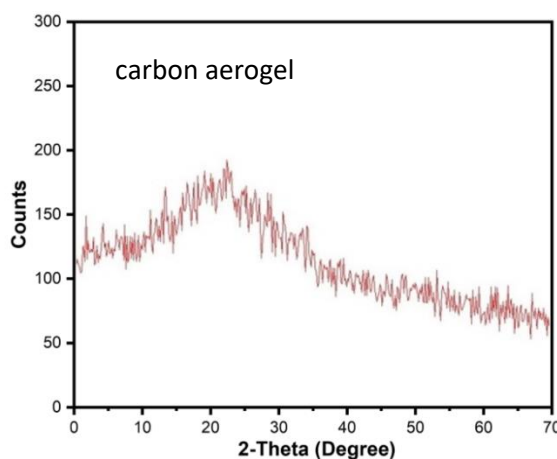


Figure 6. XRD pattern of the porous carbon CX700.

Physicochemical characterization of the materials

Table 1. Surface characterization of the CX700 sample.

S_{BET} , m^2/g	S_{mi} , m^2/g	S_{out} , m^2/g	S_{BJH} , m^2/g	V_{total} , cm^3/g	V_{mi} , cm^3/g	V_{BJH} , cm^3/g	d_{TB} , \AA
801.97	259.63	211.34	221.43	0.14	0.070	0.11	6.96

The determination of the specific surface area and capillary dimensions of the porous carbon materials was conducted using nitrogen isothermal adsorption, followed by employing the BET equation. The nitrogen adsorption-desorption isothermal curve of the sample CX700 is depicted in Figure 7. Table 1 presents a summary of the following parameters: specific surface area (S_{BET}), micropore area (S_{mi}), outer area (S_{out}), mean pore area (S_{BJH}), total volume (V_{total}), micropore volume (V_{mi}), average pore volume (V_{BJH}), and average pore size (d_{TB}).

Time to reach adsorption equilibrium

Figure 8 illustrates the effect of material contact time on methylene blue adsorption capacity. The time required to achieve adsorption equilibrium is a crucial parameter in the field

of adsorption research. The present study aimed to explore the effect of contact time on the adsorption capacity of a porous carbon sample (referred to as model CX700) activated with NaOH at a temperature of 700 °C.

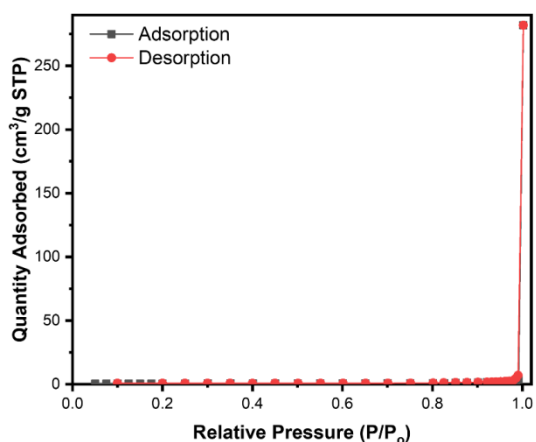


Figure 7. Isothermal nitrogen adsorption-desorption path of porous carbon.

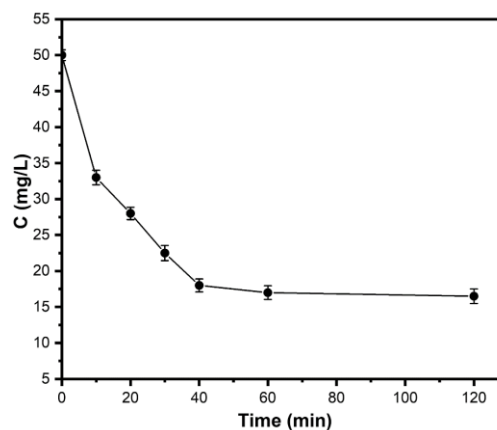


Figure 7. Studied contact time between dye and material.

From the aforementioned graph, the adsorption behavior of a porous carbon material derived from RF is presented. It is evident that this material has the ability to adsorb MB rapidly within 10 - 40 minutes. Subsequently, the adsorption rate steadily decreases, eventually reaching a state of equilibrium after 60 minutes. The rapid occurrence of the adsorption process can be attributed to the presence of a porous carbon material derived from RF, which possesses a substantial surface area and capillary dimensions that enhance the diffusion of MB molecules into the adsorption sites within the material. In order to achieve full establishment of the equilibrium state, the equilibrium time of 120 minutes was chosen for following investigations on adsorption.

Effect of pH

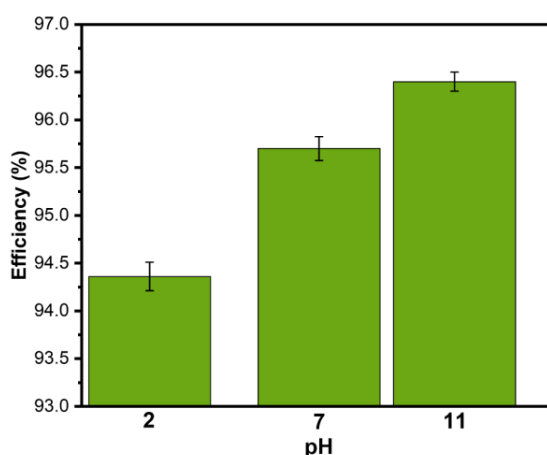


Figure 8. Adsorption efficiency corresponding to various pH levels.

It has been established that the adsorption capacity of methylene blue is influenced by factors such as surface area and the interaction between adsorbed molecules and the adsorbent

surface, mostly through electrostatic attraction. The effect of pH on the adsorption efficacy of porous carbon materials was investigated in three distinct settings: acidic, neutral, and alkaline environments. For the purpose of adsorption research, three specific pH values (2, 7, and 11) were chosen. The outcomes of this research are depicted in Figure 9.

According to the data presented in Figure 9, it can be observed that CX700 exhibits a notable level of effectiveness in adsorbing methylene blue across various pH conditions. Specifically, the adsorption efficiencies of the CX700 sample are found to be 94.36 % in acidic (pH 2), 95.70 % in neutral (pH 7), and 96.40 % in basic (pH 11) environments. In an acidic environment characterized by low pH levels, it is possible for the tertiary amine groups within the methylene blue molecule to undergo protonation. This protonation event can subsequently diminish the capacity of methylene blue to be adsorbed by the material in question. There is no substantial difference in the effectiveness of adsorption in alkaline and neutral conditions. Hence, in order to enhance practical applicability, additional investigation is being conducted on these material systems at a pH value of 7.

Adsorption isotherm

The investigation of adsorption equilibrium isotherm is conducted in order to determine the adsorption parameters, with the maximum adsorption capacity being determined based on the Langmuir equation. The adsorption process was conducted under controlled conditions, specifically at a pH of 7 and a temperature of 25 °C, using methylene blue with an initial concentration of 50 mg/L. The present study used C_e/q_e and C_e regression analysis to investigate the Langmuir model. The findings are depicted in Figure 10.

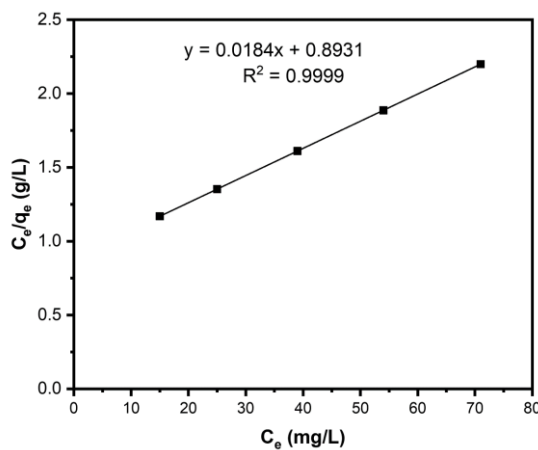


Figure 9. Linear Langmuir isotherm for adsorption of MB onto CX700.

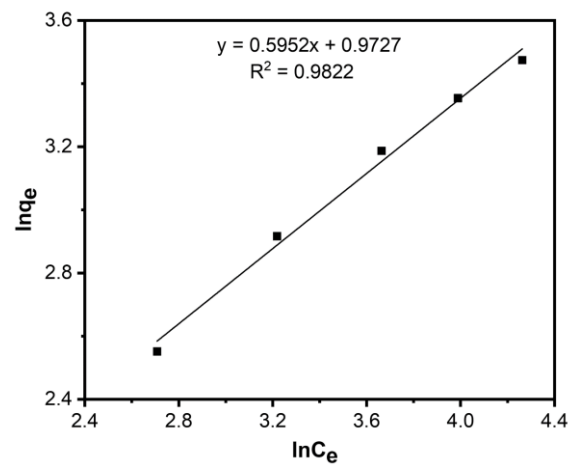


Figure 11. Linear Freundlich isotherm for adsorption of MB onto CX700.

The results show that the lines representing the dependence of C_e/q_e on equilibrium concentration C_e are linear with a correlation coefficient of $R^2 = 0.9999$. Thus, the experimental equilibrium data follow the Langmuir isotherm for the adsorption of MB onto CX700. From regression analysis, we can calculate the coefficients of the equations as follows: Langmuir equation: $\ln C_e/q_e = 0.0184C_e + 0.8931$; Freundlich equation: $\ln q_e = 0.5952 \ln C_e + 0.9727$

From Equations (3) and (4), the parameters for Langmuir and Freundlich were determined and the results are summarized in Table 2.

Table 2. Parameters for Langmuir and Freundlich isotherms.

Parameter	Langmuir	Freundlich
K_L	0.0206	-
K_F	-	2.6450
q_m	54.35	
n	-	1.6801
R^2	0.9999	0.9822

Based on the data presented in Table 2, it is evident that the correlation coefficient of the Langmuir model is close to 1. This suggests that the adsorption process of MB on the CX700 material adheres to the Langmuir isotherm model.

For the Freundlich model, there is a high linear regression coefficient, showing that the MB adsorption process follows the Freundlich isotherm adsorption equation, the adsorption process is reversible, the adsorption energy on the surface is not a uniform and the CX700 material can adsorb in multiple layers [20].

Table 3. Adsorption capacity of MB onto CX700 sample and other materials.

Material	Structure	q_m (mg/g)	Ref.
MCSG60	Silica gel	200.00	[5]
Graphene	-	153.85 - 204.08	Error! Reference source not found.
Graphene/c-MWCNT	-	190.90	Error! Reference source not found.
Activated carbon (FCBAC)	-	47.62	Error! Reference source not found.
Biomass activated carbon	-	33.00	Error! Reference source not found.
CX700	RF	54.35	Present

By conducting a comparison between the maximum adsorption capacity of methylene blue exhibited by the carbon materials synthesized in this study and those reported in recent literature (as presented in Table 3), it becomes evident that porous carbon materials, specifically carbon aerogels, possess promising characteristics for applications in the treatment of methylene blue. This is primarily attributed to their substantial surface area and extensive porous structure.

4. CONCLUSIONS

The present study aimed to assess and analyze the adsorption effectiveness of commercially produced porous carbon materials in the context of environmental remediation, specifically for the removal of methylene blue from aqueous solutions. The optimal adsorption conditions for methylene blue colorant are given as follows: At a pH value of 7 and a concentration of 50 mg/L, the adsorption effectiveness of the colorant exceeds 95 %. The adsorption process adheres to the Langmuir and Freundlich equations. The porous carbon material (CX700) exhibits a maximum adsorption capacity of 54.35 mg/g for methylene blue.

The findings of the study indicate that porous carbon materials derived from RF plastic possess promising prospects for utilization in environmental remediation. This is primarily attributed to their substantial surface area, significant porous volume, and notably, their capacity for large-scale production using readily available ingredients. The utilization of waste plastic sources contributes to environmental protection.

Acknowledgements. This section devotes to acknowledge the supports by agency and/or individual, and statement about any conflict of interest. *e.g.*: The research funding from Basic Science Research Program through the National Research Foundation of Korea (NRF) (Grant number: 2010-0008860) was acknowledged.

CRedit authorship contribution statement. Do Dinh Trung: Investigation, Funding acquisition. Vu Dinh Thao: Formal analysis. Nguyen Ngoc Linh: Formal analysis. Nguyen Duc Trung: Formal analysis, Supervision. Nguyen Ngoc Tue: Methodology, Supervision.

Declaration of competing interest. The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

REFERENCES

1. Ma Thi Van Ha - Research on the adsorption capacity of methylene blue and methylene orange on modified laterite materials, Master's thesis, Thai Nguyen, 2015 (Vietnamese).
2. Al-Muhtaseb S. A., Ritter J. A. - Preparation and properties of resorcinol-formaldehyde organic and carbon gels, *Advanced Materials* **15** (2) (2003) 101-114. doi.org/10.1002/adma.200390020.
3. Chaukura N., Murimba E. C. and Gwenzi W. - Sorptive removal of methylene blue from simulated wastewater using biochars derived from pulp and paper sludge, *Environmental Technology & Innovation* **8** (2017) 132-140. doi.org/10.1016/j.eti.2017.06.004.
4. Sharma C. S., Patil S., Saurabh S., Sharma A., Venkataraghavan R. - Resorcinol-formaldehyde based carbon nanospheres by electro spraying, *Bulletin of Materials Science* **32** (3) (2009) 239-246. doi.org/10.1007/s12034-009-0036-6.
5. Gómez J. M., Galán J., Rodríguez A., and Walker G. M. - Dye adsorption onto mesoporous materials: pH influence, kinetics and equilibrium in buffered and 140 saline media, *Journal of Environmental Management* **146** (2014) 355-361. doi.org/10.1016/j.jenvman.2014.07.041.
6. Tan Y., Wang X., Xiong F., Ding J., Qing Y., Wu Y. - Preparation of lignin-based porous carbon as an efficient absorbent for the removal of methylene blue, *Industrial Crops and Products* **171** (2021) 113980. doi.org/10.1016/j.indcrop.2021.113980.
7. Somsesta N., Sricharoenchaikul V., Aht-Ong D. - Adsorption removal of methylene blue onto activated carbon/cellulose biocomposite films: Equilibrium and kinetic studies,

- Materials Chemistry and Physics **240** (2020) 122221. doi.org/10.1016/j.matchemphys.2019.122221.
8. Vadivelan V., Kumar K. V. - Equilibrium, kinetics, mechanism, and process design for the sorption of methylene blue onto rice husk, *Journal of colloid and interface science* **286** (1) (2005) 90-100. doi.org/10.1016/j.jcis.2005.01.007.
 9. Utomo H. D., Phoon R. Y. N., Shen Z., Ng L. H., Lim Z. B. - Removal of Methylene Blue Using Chemically Modified Sugarcane Bagasse, *Natural Resources* **06** (04) (2015) 209-220. doi:10.4236/nr.2015.64019.
 10. Zhou C., Wu Q., Lei T., Negulescu I. I. - Adsorption kinetic and equilibrium studies for methylene blue dye by partially hydrolyzed polyacrylamide/cellulose nanocrystal nanocomposite hydrogels, *Chemical Engineering Journal* **251** (2014) 17-24. doi:10.1016/j.cej.2014.04.034.
 11. Romero-Anaya A. J., Ouzzine M., Lillo-Ródenas M. A., Linares-Solano A. - Spherical carbons: Synthesis, characterization and activation processes, *Carbon* **68** (2014) 296-307. doi.org/10.1016/j.carbon.2013.11.006.
 12. Baghel A., Singh B., Prasad G. K., Pandey P., Gutch P. K. - Preparation and characterization of active carbon spheres prepared by chemical activation, *Carbon* **49** (14) 4739-4744. doi.org/10.1016/j.carbon.2011.06.080.
 13. Tripathi N. K., Sathe M. - Pilot scale production of activated carbon spheres using fluidized bed reactor and its evaluation for the removal of hexavalent chromium from aqueous solutions, *Journal of the Institution of Engineers (India)* **98** (2) (2017) 141-147. doi.org/10.1007/s40034-017-0100-5.
 14. Tripathi N. K. - Porous carbon spheres: Recent developments and applications, *AIMS Materials Science* **5** (5) (2018) 1016-1052. doi.org/10.3934/matricsci.2018.5.1016.
 15. Mei Li, Xiaoqing Chang, Xianlong Han, Wenchao Yin, Manman Ren - Resorcinol-formaldehyde resin based porous carbon materials with yolk-shell structure for high-performance supercapacitors, *Synthetic Metals* **219** (2016) 67-75. doi.org/10.1016/j.synthmet.2016.05.011.
 16. Nakagawa H., Watanabe K., Harada Y., and Miura K. - Control of micropore formation in the carbonized ion exchange resin by utilizing pillar effect, *Carbon* **37** (9) (1999) 1455-1461, 1999. doi.org/10.1016/S0008-6223(99)00008-1.
 17. Cenens J., Schoonheydt R. A. - Visible spectroscopy of methylene blue on hectorite, laponite B, and barasym in aqueous suspension, *Clay and Clay Minerals* **36** (3) (1988) 214-224. doi.org/10.1346/CCMN.1988.0360302
 18. Jung C. H., Lee H. Y., Moon J. K., Won H. J., Shul Y. G. - Electrosorption of uranium ions on activated carbon fibers, *J. Radioanal. Nucl. Chem.* **287** (3) (2011) 833-839. doi.org/10.1007/s10967-010-0848-2
 19. Kim J., Lee H. Yeon J. W., Jung Y., Kim J. - Removal of uranium (VI) from aqueous solutions by nanoporous carbon and its chelating polymer composite, *Journal of Radioanalytical and Nuclear Chemistry* **286** (1) (2010) 129-133. doi.org/10.1007/s10967-010-0624-3.
 20. Myers A. L. - Thermodynamics of adsorption in porous materials, *AIChE Journal* **48** (1) (2002) 145-160. doi.org/10.1002/aic.690480115.
 21. Liu T., Li Y., Du Q., Sun J., Jiao Y., Yang G., Wang Z., Xi Y., Zhang W., Wang K., Zhu H. and Wu D. - Adsorption of methylene blue from aqueous solution by graphene.

- Colloids and Surfaces B: Biointerfaces **90** (2012) 197-203. doi.org/10.1016/j.colsurfb.2011.10.019.
22. Sui Z., Meng Q., Zhang X., Ma R. and Cao B. - Green synthesis of carbon nanotube - graphene hybrid aerogels and their use as versatile agents for water purification, Journal of Materials Chemistry **22** (18) (2012) 8767-8771. doi.org/[10.1039/C2JM00055E](https://doi.org/10.1039/C2JM00055E).
 23. Pathania D., Sharma S. and Singh P. - Removal of methylene blue by adsorption onto activated carbon developed from Ficus carica bast, Arabian Journal of Chemistry **10** (S1) (2017) 1445-1451. doi.org/10.1016/j.arabjc.2013.04.021.