ABSTRACT: Three-dimensional porous carbon was fabricated using lotus leaves as a renewable precursor. The as-synthesized carbon had a high surface area (3601 m²/g), suitable O−N−S self-doping, and three-dimensional (3D) architecture with interconnected micro/meso/macropores, together with proper pore size distribution. Consequently, these admirable features endowed porous carbon as a superadsorbent for dye removal with ultrahigh adsorption capacity for rhodamine B (9444.39 mg/g) and reliable cyclability (>97% capacitance retention after 10 cycles). The adsorption of dye onto the as-prepared carbon was a spontaneous endothermic process and followed the pseudo-second-order kinetic model and the Langmuir isotherm model. The π−π stacking, hydrogen bond, and acid−base interactions were proposed to mainly account for the combination of the adsorbate and the adsorbent. Overall, these values indicated the high-performance biomass-derived carbon as a dye adsorbent and may boost the large-scale production and application of 3D hierarchical porous carbon with heteroatom doping in the field of wastewater treatment.

1. INTRODUCTION

With the express growth of the world economy and the fast expansion of industrial production, the deterioration of environment continually accelerates, particularly, water pollution including the contamination of surface and underground water resources. Because of the extensive application of dyes in paper, plastic, rubber, textile, leather, cosmetic, glass, and pharmaceutical industries, the discharge of dye-containing industrial sewage not only leads to rise of toxicity and chemical oxygen demand but also weakens light penetration, greatly retarding the photosynthetic phenomena of the hydrophyte and the growth of microbes and hence bringing about the accumulation of impurities in water.1−6 It is easily confirmed whether the water is contaminated by industrial dye or not because of the visibility of the dye even at ~0.005 ppm.7 Among dyestuffs, rhodamine B (RhB), as a typical cationic dye, has carcinogenic and mutagenic influences on humans and animals. Therefore, the rapid and efficient removal of RhB from wastewater has attracted increasing attention over the past years.

Adsorption technology is widely used to purify dye-containing wastewater because of its low cost and easy handling characteristics. Porous carbon is regarded as a prospective adsorbent for the adsorption of various organic compounds because of its large surface area, well-developed porous structure, excellent chemical stability, and easy regeneration.8−13 Particularly, owing to its abundance, being CO₂ neutral, and fast growth, biomass-derived carbon has been proven to be an attractive alternative for dye adsorption.10,14,15 To improve the adsorption performance of carbon materials for organic dyes, many researchers have spared no effort to synthesize carbon-based adsorbents with the properties mentioned below: (1) high surface area to offer abundant adsorption sites; (2) three-dimensional (3D) hierarchical porous structure with interconnected micro/meso/macropores to boost dye molecule transportation by shortening the diffusion pathway and enhancing the mechanical stability as well; and (3) heteroatom doping (i.e., O, N, S, and P) to enhance the wettability to increase pore accessibility and electrostatic interaction between the adsorbent and the adsorbate via hydrogen bond or/and acid−base interactions to consolidate the adsorption of adsorbate molecules onto the adsorbent surface.16−20 So far, the carbon materials with these features were usually synthesized by applying a certain amount of hard template for the production of hierarchical porous carbon and applying nitrogen-, sulfur- and phosphorus-containing chemical reagents for the doping of heteroatoms...
The small mass loss at the temperature range of 200°C to 600°C in lotus leaves, including hemicellulose, cellulose, lipid, protein, and lignin, is due to the complex compositions of these components. Consequently, it would be a significantly promising way to directly prepare hierarchical porous carbon with heteroatom self-doping from renewable biomass as the precursor without the utilization of any other reagents.

The lotus root is a typical and delicious food and is widely planted in China. Consequently, abundant lotus leaves are generated every year, but most of them are discarded as waste and rot in water, leading to water pollution. However, the apparent natural features of lotus leaves afford potential advantages for the preparation of hierarchical porous carbon doped with heteroatoms: (1) richness in metal salts or high ash content, which could serve as natural hard templates for the formation of 3D architecture; (2) high contents of protein and lipid, which provide the chance of self-doping with O, N, and S heteroatoms. Unfortunately, the production of lotus leaves-based carbon and its application as adsorbent for dye removal remain unexplored.

In this work, 3D hierarchical porous carbon was fabricated from lotus leaves and applied for the removal of RhB from wastewater. The as-prepared carbon possesses a high surface area of 3601 m^2/g, a good level of O−N−S self-doping, and interconnected multipores with proper pore size distribution. These unique features lead to an ultrahigh adsorption capacitance of 9444.39 mg/g with outstanding cycling stability (>97% capacitance retention over 10 cycles). The nature of RhB adsorption onto the resultant carbon was a spontaneous endothermic process and fitted the pseudo-second-order kinetic model and the Langmuir isotherm model best. The adsorption mechanism of RhB onto the as-prepared carbon was also investigated. Thus, lotus leaves, as an abundant and renewable biomass, are a significantly promising resource for the production of superadsorbents for the purification of dye-containing wastewater.

2. RESULTS AND DISCUSSION

2.1. Material Characterizations. Figure 1 shows the thermogravimetric–mass spectrometry (TG–MS) analysis results of lotus leaves. As shown in Figure 1a, there are several weight loss peaks, which was due to the complex compositions in lotus leaves, including hemicellulose, cellulose, lipid, protein, lignin, and tannin. The small mass loss at the temperature below 200°C was attributed to the dehydration of lotus leaves and ended very quickly. It was noteworthy that the major weight loss occurred in the range of 200°C to 600°C by giving out volatiles, such as H_2O, CO, CO_2, O_2, H_2, and CH_4 (shown in Figure 1b), resulting from the thermal decomposition of cellulose, hemicelluloses, lipid, protein, and lignin in lotus leaves. Above 600°C, two slight mass loss peaks were observed, so most of the volatiles were released before the temperature of 600°C.

The micromorphology of the as-prepared lotus leaves-derived hierarchical porous carbon (LLHPC) was first detected by scanning electron microscopy (SEM). As depicted in Figure 2a−c, LLHPC exhibited a 3D porous structure with interconnected pores, which was attributed to KOH chemical activation process via C + 6KOH → K_2CO_3 + 2K + 3H_2, K_2CO_3 → K_2O + CO_2, K_2CO_3 + 2C → 2K + 3CO, and K_2O + 2C → 2K + CO with the successive post-treatment of HCl.28,29 It is noticeable that the abundant interconnected pores and open cavities obviously promoted the rapid diffusion/transfer from liquid to solid and the adsorption of dye molecules onto LLHPC. The X-ray diffraction (XRD) patterns of the as-obtained LLHPC before and after dye adsorption are presented in Figure 3. The broad and weak reflections located at about 23° and 43° were assigned to the (002) and (100) planes of the graphite carbon, which were the characteristic signals of the turbostratic carbon structure.30,31

Obviously, the ultrastrong intensity in the small-angle region (2θ < 20°) indicated the presence of a great number of micro pores in LLHPC.31 Thus, LLHPC is the amorphous carbon material with 3D porous architecture.

X-ray photoelectron spectroscopy (XPS) was further applied to determine the chemical composition and states of surface atoms of the as-synthesized material, and the results are displayed in Figure 4. The contents of C, O, N, and S in LLHPC were 86.49, 10.20, 1.44, and 1.78 at %, respectively. The C 1s spectrum was deconvoluted into four peaks of C−C (~284.8 eV), C−N (~285.7 eV), C==O (~287.1 eV), and COOR (~290 eV).32,33 The high-resolution spectrum of O 1s demonstrated three kinds of oxygen-containing functional groups of C==O (~532.2 eV), C−O (~533.2 eV), and COOR (~534.4 eV).34 The N 1s spectrum was fitted into two peaks at ~399 and ~402 eV, indicating the presence of pyridine and quaternary.35 The S 2p spectrum revealed the existence of sulfone bridges (C==SO−C) centered at ~169.0 and ~170.2 eV.36 These results demonstrated the successful self-doping of O, N, and S heteroatoms in the as-prepared carbon material. It was known that the doping of O, N, and S heteroatoms and the

![Figure 1](https://dx.doi.org/10.1021/acsomega.0c02021)
related polar groups not only enhanced the hydrogen bond/acid–base interaction between the adsorbent and dye molecules but also improved the wettability, thereby increasing the pore accessibility and accelerating the diffusion of adsorbate molecules.

The specific surface area and pore property of the as-obtained LLHPC were measured by N$_2$ adsorption and desorption analyses at $-196\, ^\circ$C. Figure 5a,b depicts the N$_2$ adsorption–desorption isotherm and pore size distribution. It can be seen from Figure 5a that LLHPC possessed an ultrahigh adsorption capacitance and showed the combined characteristics of type I and IV isotherms with a hysteresis loop. The dramatic increase in the volume adsorbed at a relative pressure <0.05 indicated the existence of a great number of micropores, in agreement with the analyses of SEM and XRD. The observation of the hysteresis loop resulting from the high capillary effect at a relative pressure in the range of 0.4–0.8 suggested the presence of a mesopore structure. Moreover, the slight increase at a relative pressure >0.8 demonstrated the presence of a number of macropores. Additionally, the pore size distribution in Figure 5b also revealed the microporous and mesoporous structures with the presence of macropores. Thus, the hierarchical porous carbon material was successfully synthesized, which benefited the fast adsorption and diffusion/transportation of adsorbate molecules.$^{15,16}$ Consequently, LLHPC was confirmed to possess a high surface area of 3601 m$^2$/g, a large pore volume of 2.44 cm$^3$/g (micropore volume of 0.93 cm$^3$/g), and an average pore diameter of 2.56 nm. Overall, LLHPC had a high surface area, appropriate O–N–S heteroatom doping, and 3D architecture with multi-connected micro/meso/macropores with proper pore size distribution. These admirable properties may make the as-prepared carbon material a significant superadsorbent for the efficient removal of dye from wastewater.

2.2. Dye Adsorption Capability. The influences of adsorption parameters (e.g., contact time, pH, initial concentration, and temperature) were studied at the beginning, and the results are shown in Figure 6. The impact of contact time on the adsorption capacitance is depicted in Figure 6a,b, which was carried out by the mixture of 10 mg of LLHPC and 100 mL of RhB solution (200 mg/L) at room temperature and pH value of 7. $q_t$ drastically increased in the first 10 min and then reached equilibrium with $q_e$ of 1990.7 mg/g and the dye removal percentage of 99.5%, suggesting the fast and efficient purification of RhB-containing wastewater. It was attributed to the 3D interconnected hierarchical porous structure with suitable pore size distribution boosting the diffusion of dye molecules, high surface area, and good level of interaction with the adsorbate.
O–N–S heteroatom doping, offering immense active sites for RhB adsorption. The relation between the adsorption capability and the initial pH value is displayed in Figure 6c, performed by immersing 10 mg of LLHPC in 100 mL of 200 mg/L RhB solution at room temperature and pH value in the range of 3–11. The increase in pH value showed a favorable impact on the enhancement of adsorption rate; the rise in pH increased the negative charge of LLHPC and enhanced the electrostatic interaction between LLHPC and RhB molecules (cationic dye). Noticeably, the change of pH just led to a small change of the adsorption rate and prolonged the time to adsorption equilibrium but showed no effect on the adsorption capacitances at equilibrium, indicating significant potential of the adsorbent toward RhB in a wide pH range. The effects of initial concentration and temperature are illustrated in Figure 6d, which were examined at different temperatures (25–60 °C) and a pH value of 7 by using 10 mg of adsorbent and 100 mL of RhB solution with different initial concentrations (200, 400, and 600 mg/L). It was found that both the initial concentration and the temperature showed beneficial impacts on the adsorption capacity. Two reasons accounted for the results. First, the growth in initial concentration enhanced the driving force to overcome the mass transfer resistance of the adsorbate between the liquid and the solid adsorbent. Second, the adsorption of RhB onto LLHPC was an endothermic process.

For the adsorption kinetic assessment, the fitting linear plots of RhB adsorption performance of LLHPC as a function of contact time are displayed in Figure 7 and the linked parameters are listed in Table 1. It was found that the
pseudo-second-order kinetic model described best the adsorption of RhB onto as-prepared carbon with the highest $R^2$ value of 1.0, and the calculated adsorption capacitance ($q_e$) of 1991.13 mg/g was very close to the experimental value. Therefore, the adsorption of RhB onto LLHPC allowed the pseudo-second-order kinetic model, indicating the natural adsorption process of chemisorption. It was attributed to O–N–S heteroatom doping and the related functional groups.

For the adsorption isotherm evaluation, the fitting linear plots of experimental data as a function of equilibrium concentration are displayed in Figure 8 and the associated parameters are summarized in Table 2. It can be seen that the adsorption process of RhB onto LLHPC followed the Langmuir isotherm model best with the highest $R^2$ of 0.9925, revealing the uniformity of the adsorption sites over LLHPC and the monolayer adsorption of RhB molecules with no transmigration. The maximum monolayer adsorption capacity ($Q_{max}$), determined by the application of the Langmuir isotherm model was a new record of 9444.39 mg/g, highest among those reported in the literature,13,18,37 which resulted from the high surface area, hierarchical porous structure, and O–N–S codoping. Equilibrium parameter ($R_L$) was calculated from $R_L = \frac{1}{1 + K_L C_e}$ to investigate the feasibility of adsorption onto the adsorbent, and $R_L$ (0.004–

Figure 5. (a) N$_2$ adsorption–desorption isotherm and (b) pore size distribution of as-obtained LLHPC.

Figure 6. Effects of (a,b) contact time, (c) initial pH, and (d) initial concentration and adsorption temperature on the adsorption performance of as-prepared LLHPC.
0.018) was in the range of $0 \sim 1$, indicating the favorable nature of adsorption process onto LLHPC.\(^{38,39}\) For the adsorption thermodynamic study, the linear plots of $\ln K_c$ against $1/T$ are shown in Figure 9 and the associated parameters, calculated from the linear regression analysis, are summarized in Table 3. The positive values of $\Delta H^\circ$ and $\Delta S^\circ$ indicated the endothermic nature of the adsorption process and the increase of system randomness, whereas the negative value of $\Delta G^\circ$ demonstrated the spontaneous nature of RhB adsorbed onto LLHPC. As a result, the porous carbon derived from lotus leaves exhibited ultraexcellent adsorption rate and adsorption capacitance. Hence, it is an efficient and low-cost alternative superadsorbent for wastewater treatment, especially for dye removal.

The reusability stabilization is an important factor for the practical application of LLHPC in the purification of wastewater. It can be seen from Figure 10 that the adsorption capacity of LLHPC slightly decreased from 7798.9 to 7609.9 mg/g after four cycles and then remained stable at $\sim 7600$ mg/g with the capacitance retention of $>97\%$. Consequently, the as-synthesized LLHPC exhibited outstanding recyclability for the purification of dye-containing wastewater.

The Fourier transform infrared (FTIR) spectrum was applied to comprehend the adsorption mechanism of RhB molecules onto LLHPC.\(^{40}\) Figure 11 displays the FTIR spectra of pure RhB and LLHPC before and after the adsorption of dye molecules. The FTIR spectrum of unused LLHPC powder offered the characteristics of organic groups below: the signal at $\sim 3430$ cm$^{-1}$ belonged to the stretching vibration of O$\sim$H or N$\sim$H;\(^{40}\) the peak at $\sim 2930$ cm$^{-1}$ was attributed to the C$\sim$H stretching vibration of methyl groups;\(^{41}\) the band at 1628 cm$^{-1}$ corresponded to the stretching vibrations of C=C, C=O, and C=N;\(^{42}\) the narrow band at $\sim 1400$ cm$^{-1}$ was assigned to the C--N stretching vibration;\(^{40}\) the adsorptions from 1330 to 940 cm$^{-1}$ were identified as the response of C=N, C=O, and S=O stretching vibrations; and the peak at 1048 cm$^{-1}$ was attributed to the C--O--C stretching vibration. These results are consistent with those of the XPS analyses. The FTIR spectrum of spent LLHPC displayed the apparent combined adsorption information of LLHPC and RhB, demonstrating the successful adsorption of RhB onto LLHPC. It was evident that the surface of as-prepared LLHPC located abundant oxygen-functional groups (e.g., O--H, C=O, and C=O) and $\pi--\pi$ structures, and the structure of the RhB molecule exhibited a

![Figure 7](fig7.png)

**Figure 7.** Linear plots of (a) $\ln(q_e - q_t)$ as a function of $t$, (b) $t/q_t$ as a function of $t$, (c) $q_t$ as a function of $t^{0.5}$, and (d) $q_t$ as a function of $t \ln t$ for the dye adsorption process onto LLHPC.

| Table 1. $R^2$ and Constants for the Different Adsorption Kinetics Models |
|-----------------------------|-----------------------------|-----------------------------|-----------------------------|-----------------------------|
| models                      | pseudo-first-order          | pseudo-second-order         | intraparticle               | elovich                     |
| $R^2$                       | 0.9847                      | 1.0                         | 0.9784                      | 0.8861                      |
| constants                   | $k_1$ 0.5519 min$^{-1}$     | $k_2$ 2.1 x 10$^{-3}$ g/(mg-min) | $k_1$ 767.02 mg/(g·min$^{0.5}$) | $\beta$ 2.38 x 10$^{-3}$ g/mg |
|                            | $q_e$ 1925.88 mg/g          | $q_e$ 1991.13 mg/g          | $q_e$ 1991.58 mg/g          | $\alpha$ 5923.82 mg/(g·min) |

0.018) was in the range of $0 \sim 1$, indicating the favorable nature of adsorption process onto LLHPC.\(^{38,39}\)

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hydroxyl group and an aromatic backbone. As a result, the combination of the adsorbate and the adsorbent was concluded to occur via π−π stacking and the formation of hydrogen bond. In addition, LLHPC was doped with proper level of O, N, and S heteroatoms, which can act as the base to provide electrons or accept protons, and the organic ammonium cations in the RhB molecule can serve as acid to accept electrons or provide protons. Thus, the acid−base interaction also made a contribution to the adsorption of RhB onto LLHPC. The ultraexcellent adsorption performance resulted from π−π stacking, hydrogen bond, and acid−base interactions.

3. CONCLUSIONS

3D hierarchical porous carbon was successfully derived from lotus leaves and possessed a significantly high surface area of 3601 m²/g, appropriate O−N−S self-doping, and 3D architecture with interconnected micro/meso/macropores. These advantageous features afforded ultraexcellent adsorption capacity as high as 9444.39 mg/g. The adsorption of dye onto LLHPC was a spontaneous endothermic process and fitted the pseudo-second-order kinetic model and Langmuir isotherm models.
The adsorption mechanism was mainly attributed to the π−π stacking, hydrogen bond, and acid−base interactions. The superior adsorption performance closely originated from the natural properties of lotus leaves, including high contents of proteins and lipid for the self-doping of different heteroatoms and richness of metal salts, serving as natural templates for the generation of 3D architecture. The utilization of lotus leaves as a renewable precursor would pave the way for cost-efficient production of heteroatoms self-doped 3D hierarchical porous carbon.

4. EXPERIMENTAL SECTION

4.1. Materials. Lotus leaves were collected from the campus of Anhui Polytechnic University in Wuhu (Anhui province, China). They were first washed with distilled water to remove dust particles and then dried at 110 °C, crushed, and sieved to obtain a uniform grain size in the range of 0.2−0.5 mm. The proximate [according to the Chinese standard (GB/T 28731−2012)] and ultimate analyses of the raw material are illustrated in Table 4. The ash and nitrogen contents of lotus leaves were 13.63 and 3.15%, which was due to the composition rich in metal salts and protein. Table 5 lists the ash composition; the major components in ash were metallic oxides, such as CaO and K2O, which can function as hard templates to generate a porous structure.

4.2. Preparation of Porous LLHPC. Lotus leaves were first pyrolyzed in a fixed bed reactor under N2 protection. The temperature was continuously increased to 600 °C at a heating rate of 3 °C/min and then kept for 1 h. For the activation process, the resultant powder from the previous pyrolysis process was ground with KOH at the mass ratio of 1: 4. Subsequently, the mixed powder was placed in a tube furnace and heated to 800 °C at a rate of 2 °C/min and maintained for 1 h under a N2 atmosphere. Finally, the as-synthesized powder was cooled to room temperature and washed with 1 M HCl solution and distilled water until neutral and dried at 110 °C overnight to obtain LLHPC.

4.3. Characterizations. The elemental analyses in the lotus leaves were performed by an elemental analyzer (Elementar, vario MACRO CHN). An X-ray fluorescence (XRF) analyzer (Panalytical, AXIOS) was used to analyze the compositions of the ash derived from lotus leaves. The thermal behavior of lotus leaves was evaluated with a thermal analyzer (Hitachi, TG/DTA 7300). The samples were heated up to 900 °C in an Ar atmosphere at a heating rate of 5 °C/min. Brunauer−Emmett−Teller (BET) surface area, pore volume, and pore size of the activated carbons were analyzed by N2 adsorption/desorption measurements (Micrometric, model ASAP2020). The sample was first degassed at 250 °C under vacuum overnight, and then the analysis was carried out by using N2 as an adsorbate gas at −196 °C. BET surface area and pore size distribution were determined by the BET theory and the nonlocal density functional theory, respectively.

4.4. Adsorption Test. For the dye adsorption test onto as-synthesized LLHPC, batch adsorption experiments were performed. The adsorption isotherms were measured at different initial concentrations using a Langmuir−Freundlich model. The adsorption isotherms were mainly attributed to the π−π stacking, hydrogen bond, and acid−base interactions. The superior adsorption performance closely originated from the natural properties of lotus leaves, including high contents of proteins and lipid for the self-doping of different heteroatoms and richness of metal salts, serving as natural templates for the generation of 3D architecture. The utilization of lotus leaves as a renewable precursor would pave the way for cost-efficient production of heteroatoms self-doped 3D hierarchical porous carbon.

Table 3. Thermodynamic Parameters for the Adsorption of RhB Onto Lotus Leaves-Derived Carbon

| initial concentration (mg/L) | ΔH° (kJ·mol−1) | ΔS° (J·mol−1·K−1) | ΔG° (kJ·mol−1) |
|-----------------------------|--------------|------------------|--------------|
|                             | 298 K        | 313 K            | 323 K        | 333 K        |
| 200                         | −12.73       | −14.64           | −15.92       | −17.19       |
| 400                         | −12.07       | −13.43           | −14.33       | −15.23       |
| 600                         | −11.62       | −13.07           | −14.04       | −15          |

Table 4. Proximate and Ultimate Analyses of Lotus Leaves

| proximate analysis (wt %) | ultimate analysis (wt %) |
|--------------------------|--------------------------|
| moisture                 | C                        |
| ash                      | H                        |
| volatile                 | O                        |
| fixed carbon             | N                        |
| 5.08                     | 43.59                    |
| 13.63                    | 5.49                     |
| 64.62                    | 47.19                    |
| 16.67                    | 3.15                     |
|                            | 0.58                     |

Figure 10. Recycling of LLHPC for adsorption of RhB by adsorption−desorption processes.

Figure 11. FTIR spectra of pure RhB and LLHPC before and after the adsorption of dye molecules.
performed by mixing a given amount of LLHPC and RhB aqueous solution at a preset pH value and successively shaking for a certain period of time. After centrifugation for about 15 min to remove LLHPC powder, the RhB concentration remaining in the solution was measured by an ultraviolet–visible spectrophotometer (Shimadzu, UV 2700) at the wavelength of 554 nm. The impacts of contact time, pH value, initial RhB concentration, and adsorption temperature were explored.

The amount of adsorbed dye ($q_t$) onto LLHPC and dye removal percentage (removal) were determined from eqs 1 and 2, respectively.

$$q_t = \frac{(C_o - C_t) \times V}{m}$$

(1)

$$\text{removal} = \frac{C_o - C_t}{C_o} \times 100\%$$

(2)

where $q_t$ (mg/g) is the dye adsorption capacitance onto LLHPC at $t$ (min), $C_o$ (mg/L) is the initial RhB concentration, $C_t$ (mg/L) is the RhB concentration remaining in the solution at $t$ (min), $V$ (L) is the volume of RhB solution, and $m$ (g) is the mass weight of LLHPC used in experiment.

The adsorption experiments for the adsorption kinetic assessment were carried out at room temperature by immersing 10 mg of LLHPC powder in 100 mL of 200 mg/L RhB solution at a pH value of 7. The concentration of the remaining dye was measured at certain time intervals. The four kinetic models of pseudo-first-order, pseudo-second-order, intraparticle diffusion, and elovich were used to describe the relation between adsorption performance and contact time. The linear equations and the linked parameters of these kinetic models are summarized in Table 6.13,36

| models          | names                      | expressions                          |
|-----------------|----------------------------|--------------------------------------|
| kinetic         | pseudo-first-order         | $ln(q_t - q_e) = ln q_i - k_1 t$    |
|                 | pseudo-second-order        | $\frac{1}{q_t} = \frac{1}{q_i} + \frac{k_2}{k_1 t}$ |
|                 | intraparticle diffusion    | $q_t = k_3 t^{0.5}$                  |
|                 | elovich                    | $q_t = \frac{1}{\beta} ln(a_0 t) + \frac{1}{\beta} ln t$ |

where $k_1$ (min$^{-1}$), $k_2$ (g·mg$^{-1}$·min$^{-1}$) and $k_3$ (g·mg$^{-1}$·min$^{-0.5}$) represent the rate constants, $a$ (g·mg$^{-1}$·min$^{-1}$) is the initial adsorption rate, and $\beta$ (g/mg) is the desorption rate. $Q_{max}$ (mg/g) is the maximum adsorption capacity, $K$ (L/mg) and $T$ (L/g) are the constants, $n$ represents the heterogeneity of the adsorbent surface, $a_1$ (L/g) and $b_1$ (L/mg) are the constants of Temkin isotherm, $B$ is a constant, $Q_m$ (mg/g) is the saturation adsorption capacity, and $e$ is the Poayni potential.

To disclose the interaction between the adsorbate and the adsorbent, the adsorption isotherm study was performed at room temperature by mixing 10 mg of LLHPC with 100 mL of RhB solution in the range of 100–800 mg/L at pH 7. The mixture was shaken for 12 h until the adsorption equilibrium reached. The four kinds of isotherm models, namely, Langmuir, Freundlich, Temkin, and Dubinin–Radushkevich, were applied to fit the adsorption performance of RhB onto LLHPC against equilibrium concentration, and the linear equations and the associated parameters are listed in Table 6 as well.13

| models          | names                      | expressions                          |
|-----------------|----------------------------|--------------------------------------|
|                 | Langmuir                  | $Q_t = \frac{1}{1 + \frac{C_o}{Q_{max}}}$ |
|                 | Freundlich                | $ln q_t = ln K_F + \frac{1}{n} ln C_s$ |
|                 | Temkin                    | $Q_t = \frac{RT}{b_1}ln C_s + \frac{RT}{b_1}$ |
|                 | Dubinin–Radushkevich     | $ln q_t = ln Q_m - B e^2$            |

where $K_F$ (L/mg) and $K_s$ (L/g) are the constants of Temkin isotherm, $B$ is a constant, $Q_m$ (mg/g) is the saturation adsorption capacity, and $e$ is the Poayni potential.

To comprehend the nature and feasibility of RhB adsorption onto as-obtained carbon, adsorption thermodynamic evaluation was undertaken in the temperature range of 25–60 °C with different initial concentrations of 100 mL of RhB solution (200–600 mg/L) by using 10 mg of LLHPC; pH and contact time were controlled at 7 and 12 h, respectively. Thermodynamic parameters were calculated from the following equations.13

$$\Delta G^o = -RT ln K_c$$

(3)

$$K_c = \frac{C_o - C_t}{C_s}$$

(4)

where $\Delta G^o$ (J/mol), $\Delta H^o$ (J/mol), and $\Delta S^o$ (J·mol$^{-1}$·K$^{-1}$) represent the Gibbs free energy change, enthalpy change, and entropy change, respectively, $K_c$ is the equilibrium constant, and $C_s$ (mg/L) is the concentration of the remaining RhB solution at equilibrium.

In the recyclability examination, the adsorption experiment was conducted at room temperature by immersing 10 mg of LLHPC in 100 mL of RhB solution (800 mg/L) at pH 7 and successively shaking for 12 h, which was similar to the adsorption isotherm studies. The desorption experiment was performed by mixing the spent LLHPC with excess ethanol, shaking for 5 min, and successively centrifuging for 15 min to separate LLHPC. The as-separated carbon was dried in an oven for 12 h at 120 °C to remove adsorbed ethanol. The comparison of LLHPC before and after dye adsorption is shown in the Supporting Information.

Table 5. Results of XRF Analyses of the Ash from Lotus Leaves (wt %)

| CaO  | K$_2$O | P$_2$O$_5$ | Cl  | MgO  | SO$_2$ | MnO  | SiO$_2$ | Na$_2$O | Fe$_2$O$_3$ | SrO  | Al$_2$O$_3$ | Rh$_2$O | ZnO  |
|------|--------|------------|-----|------|--------|------|---------|---------|-------------|------|-------------|--------|------|
| 55.611| 18.691| 8.312      | 6.994| 3.993| 2.975  | 1.845| 0.592   | 0.443   | 0.216       | 0.147| 0.066       | 0.056  | 0.040|

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SEM images, XRD patterns, N$_2$ adsorption–desorption isotherm, and pore size distribution of as-obtained LLHPC after the adsorption of RhB molecules (PDF)
Preparation of Reduced Graphene Oxide/Poly(acrylamide) Nano-templated hydrothermal carbons for dye removal from water.

Kukk, E.; Sillanpää, A. Adsorptive removal of hazardous anionic dye mixed-component metal-organic porous material. J. Large-scale fabrication of N-doped porous carbon nanosheets for dye adsorption and supercapacitor applications. J. Mater. Sci. 2017, 52, 11201−11228.

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Notes
The authors declare no competing financial interest.

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