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Artocarpusodoratissimus peel as a potential adsorbent in environmental remediation to remove toxic Rhodamine B dye

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- ✓ water remediation,
- ✓ Rhodamine B dye,
- ✓ Regeneration

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Abstract

This research highlights the potential utilization of the skin of a locally abundant fruit, *Artocarpusodoratissimus,* also known as Tarap, for the removal of toxic Rhodamine B (RhB) dye from aqueous solution. Batch adsorption experiments carried out by varying experimental conditions indicated that the optimum conditions for adsorption of RhB on Tarap skin (TS) are 210 min shaking time and at ambient pH of 4.15. Application of experimental adsorption data on Langmuir, Freundlich and Redlich-Peterson isotherm models indicated that adsorption of RhB on the adsorbent is best described by the Langmuir model with a good maximum adsorption capacity of 131 mg g^{-1}, a value which is much higher when compared to many reported natural adsorbents. Fourier Transform Infrared spectroscopy indicates possible involvement of functional groups such as amine, alcohol and carbonyl, in the adsorption process. Adsorption under investigation was endothermic in nature. The adsorbent is resilient to both changes in ionic strength and medium pH, and could be regenerated and reused for at least four consecutive cycles.

1. Introduction

Water is a fundamental element in life, and 70% of the Earth's surface is covered by water. It may seem that the amount of water is abundant; however, only less than 1% of fresh water is available for human consumption. Therefore, water is one of the most valuable and limited resources around worldwide. In spite of water being short in adequacy, water pollution has always been a global concern as the increase in water pollutants is constantly rising. Industrial pollutants, such as dyes and heavy metals, are often inadequately treated and discharged into water, thereby polluting the usable water supply. At the same time, this greatly disrupts the ecosystem of aquatic animals and plants, as well as posing detrimental health problems to mankind. As countries become more and more developed, the demands from growing population naturally become higher. As a result, more industries have to be established to meet the increasing demand. Dealers often manufacture their products in colorful and unique packaging with the intention of attracting consumers. This results in large scale usage of dyes in the production of food, textiles, clay, paper, cosmetics, etc. Consequently, more industrial wastes, domestic wastes and other wastes are being and will continue to be dumped into the water.

One effective way that has attracted much attention in recent years is the use of adsorption technique in cleaning wastewater. Various adsorbents ranging from industrial wastes [1-2], agricultural wastes [3-5], aquatic plants [6], leaves [7] peat [8-11], fungi [12-13], bacteria [14], and many others [15-17] have been investigated for their adsorption ability toward heavy metals and dyes. However, not all of these adsorbents gave good adsorption capacity, and hence, there is still a need in search for economical and more effective adsorbents.

Artocarpusodoratissimus, locally known as Tarap in Brunei Darussalam, is available seasonally and is known to have the maximum diversity occurring in Brunei Darussalam. During the fruiting season, this fruit is fairly cheap. Both the flesh and seeds of *Artocarpusodoratissimus* are edible while the skin and core are discarded as wastes. Generally these wastes from *Artocarpus spp.* can account for more than 50% of the whole fruit [18-19]. Utilizing of these wastes as adsorbents is therefore attractive as this helps reduce waste disposal problems while at the same time converts the wastes into useful adsorbents to clean up wastewater. Several dyes and heavy metals have been successfully removed through the use of Tarap wastes such as skin [20-22], core [23] and leaf [24] as adsorbents. In fact, *Artocarpus* species, such as jackfruit [25,26], *A. altilis* (breadfruit) [27,28], *A. camansi* (breadnut) [29-31] and some *Artocarpus* hybrids [32-35], have been successfully used as adsorbents in removing dyes and heavy metals through adsorption method.

This study investigates the use of *A. odoratissimus*(Tarap) skin (TS) as a natural adsorbent to adsorb toxic Rhodamine B (RhB) cationic dye. RhB, a strong oxidizing agent, is known to have an acute toxicity with unclassifiable carcinogenic effect toward human. It can cause serious damage to eyes and is toxic to aquatic life which may cause adverse effects in their ecosystem [36]. Although RhB is not thought as a skin irritant, it could eventually bring about abrasive damage after long term exposure. Moreover, consuming products containing RhB may eventually lead to soft tissue cancer due to high carcinogenicity. RhB may also affect human's reproductive activities as well as neural activities. According to Registry of Toxic Effects of Chemical Substances (RTECS), RhB is capable of forming equivocal tumor. It has been found, through animal studies, to induce disease such as lymphoma and is poisonous to a fetus. To date, no report on the use of TS for the removal of RhB dye is available. The goal of this research is therefore to explore the ability of TS as an adsorbent for RhB with possible extension toward large scale treatment of RhB contaminated wastewater in future.

2. **Experimental**

2.1. *Materials*

Random samples of *Artocarpusodoratissimus*(Tarap) fruits were purchased from local open markets. The Tarap skin (TS) was separated from the rest of the fruit and dried in an oven at 85 °C until a constant mass was obtained. The dried TS was then blended and sieved to obtain particles of diameter < 355 µm, followed by thorough mixing. Samples prepared in this manner were used throughout the experiment.

2.2. *Chemicals and instrumentations*

Rhodamine B (RhB) dye (molecular weight 479.02 g mol^{-1} and molecular formula, $C_{28}H_{31}ClN_2O_3$) and potassium nitrate (KNO_3) were purchased from Sigma-Aldrich. Adjustment of pH was done using sodium hydroxide (NaOH) and nitric acid (HNO_3), purchased from Univar and AnalaR, respectively. A 1000 mg L^{-1} RhB dye stock solution was prepared by dissolving 1.0 g of RhB in 1 L double distilled water. From this stock solution, dye solutions of diluted concentrations were prepared. All reagents were used without further purification. Shimadzu UV-1601PC UV–Visible spectrophotometer (UV-Vis) was used to analyze the dye concentrations at its maximum wavelength of 555 nm. Functional group characterization using Shimadzu IR Prestige-21 spectrophotometer was recorded at spectral range from 4000 to 400 cm^{-1}.

2.3. *Batch experimental procedure*

Batch experiments were carried out by agitating a mixture of TS in 25.0 mL of known RhB concentration in an adsorbate:adsorbent ratio of 1:500 (mass:volume). Optimization of parameters for contact time, medium pH and ionic strength was determined using methods as described by Lim *et al*. [29].

The amount of dye adsorbed per gram of TS, q_e (mmol g^{-1}), was calculated using Equation (1).

$$q_e = V(C_o\text{-}C_e)/Mm \qquad (1)$$

whereC_0 is the initial dye concentration (mg L^{-1}), C_e is the equilibrium dye concentration (mg L^{-1}), M is the molecular mass of RhB (g mol^{-1}), V is the volume of RhB used (L), and m is the mass of TS used (g). The percentage removal of the dye is represented by Equation (2) below:

$$\text{Removal (\%)} = 100(C_o\text{-}C_e)/C_o \qquad (2)$$

2.4. *Regeneration studies*

Regeneration studies on TS were carried out using method as described by Kooh*et al*. [37]. TS that had been previously loaded with RhB dye was subjected to various treatment methods: double distilled water, 0.1 M hydrochloric acid (HCl), 0.1 M sodium hydroxide (NaOH), heating at 200 °C and no treatment, in order to evaluate if the adsorbent could be regenerated and reused.

2.5. *Error analyses*

In order to determine the best isotherm model for the adsorption of RhB dye onto TS, the experimental data was analyzed based on the correlation coefficient (R^2) value, simulation of non-linear isotherm models and error function analyses. Four error functions used in this study are the Average relative errors (ARE), Sum square error (SSE), Hybrid fractional error function (HYBRID)and the chi-square test (χ^2) as shown in equations (3) to (6), respectively. Smaller values of these error analyses indicate better curve fitting [38].

Average relative errors (ARE): $\frac{100}{n}\sum_{i=1}^{n}\left|\frac{q_{e,meas}-q_{e,calc}}{q_{e,meas}}\right|_i$ (3)

Sum square error (SSE): $\sum_{i=1}^{n}(q_{e,calc}-q_{e,meas})_i^2$ (4)

Hybrid fractional error function (HYBRID): $\frac{100}{n-p}\sum_{i=1}^{n}\left[\frac{q_{e,meas}-q_{e,calc}}{q_{e,meas}}\right]_i$ (5)

χ^2: $\sum_{i=1}^{n}\frac{(q_{e,meas}-q_{e,calc})^2}{q_{e,meas}}$ (6)

where*n* is the number of parameter and *p* is the number of data in the experiment while $q_{e,meas}$ and $q_{e,calc}$ are the experimental and calculated values, respectively.

3. **Results and discussion**

3.1. *Effects of contact time, medium pH and ionic strength*

Prior to adsorption isotherm and kinetics studies, optimization of parameters, such as shaking time, medium pH and ionic strength, was investigated. An important parameter that should be optimized in adsorption studies is the contact time, which ensures that the adsorbent-adsorbate system reaches complete equilibrium. In this study, a rapid removal of RhB by TS occurred during the first 30 min, indicating the abundance of many vacant active sites on the surface of the adsorbent, thereby allowing the dye molecules to be readily adsorbed. Thereafter, the rate of adsorption decreased and eventually reached a point of saturation. The optimum contact time required for the adsorption of RhB onto TS was determined at 210 min.

TS displayed resilience to effect of medium pH and as shown in Figure 1(A). Over the range of pH 3 to 10, only less than 10% reduction in the extent of removal was observed. Higher removal of RhB was observed at pH 3 and 4, within which RhB, having the pK_a value of 3.2, would have a sufficiently high concentration of its natural cationic form under normal temperature conditions. Increase in medium pH would make the RhB neutral or anionic form with ionized carboxylic acid group, which shows slightly weaker attraction. Further, RhB could also be present in its monomeric form within this pH range. As the monomeric form of RhB itself is large, it is not very likely to diffuse RhB adsorbate molecules/ions into micropores of TS, leaving out the possibility of intra-particle diffusion. This situation becomes even less possible at pH > 4, because the monomeric form would be converted to the dimeric form [33]. As the extent of removal is not pH dependent, RhB being in monomeric or dimeric form would not significantly affect the adsorption process, which further supports the claim that diffusion would not be a key step of mass transfer, implying that electrostatic attraction would be the principal mode of mass transfer. Further, since there is no significant pH dependence of RhB removal, no adjustment of medium pH was made in all subsequent experiments. TS shows strong removal ability toward RhB at its ambient solution pH of 4.15, as shown in Figure 1(A).

It is common to detect the presence of different ions in wastewater, and the presence of high concentration of salts could indirectly affect the extent of removal of dye molecules. Hence, the study on the effect of ions on adsorption was carried out with two different salts, namely, KNO_3 and KCl. It was found that similar extent of removal values of RhB by TS were observed in both salt media exhibiting resilience to change in ionic strength, as shown in Figure 1(B). The slight 7% reduction in the extent of removal at higher salt concentrations observed in both cases may be the result of electrostatic repulsion caused by the salt [37].

The ability of TS being relatively unaffected by both medium pH and salt concentration clearly demonstrates that TS has a strong affinity as an adsorbent for RhB dye. This is an advantage and an indication that TS could be a possible potential low-cost adsorbent for the removal of RhB dye in real life application since textile wastewater usually consists of high ionic strength due to the presence of salts.

3.2. *Adsorption isotherm and thermodynamics studies*

Three isotherm models, namely Langmuir [40], Freundlich [41] and Redlich and Peterson (R-P) [42], were used to evaluate adsorption characteristics of RhB on TS within the initial concentration range from 0 to 1000 mg L^{-1} under batch experimental conditions.

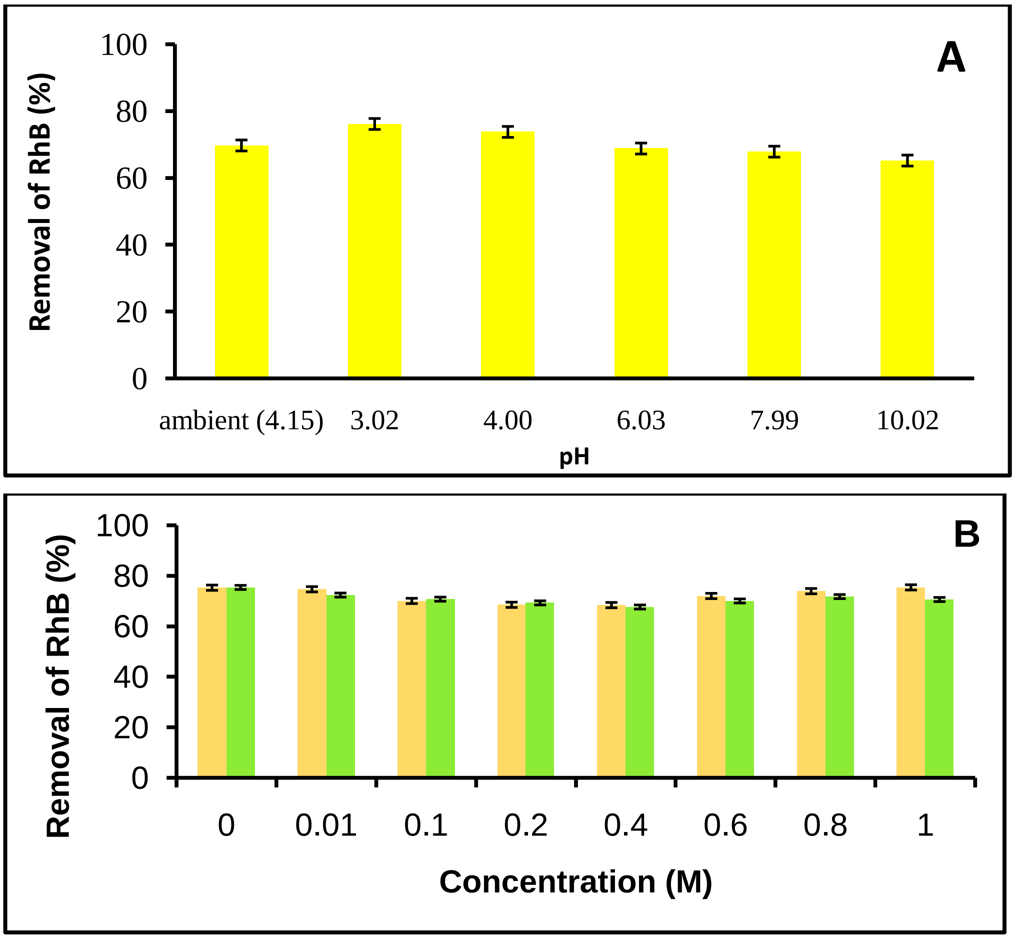

Figure 1: Effects of (A) medium pH and (B) salt concentrations using KNO_3 ( ) and KCl ( )

The Langmuir isotherm model describes the adsorption of adsorbate molecules on the surface of the adsorbent is monolayer and takes place at specific homogenous sites with identical uniformity in energy. Hence, the probability of molecules distributing on all adsorption sites should be the same. However, there is a limit to the amount of molecules that the surface can adsorb. Once their saturation point is reached, no further adsorption occurs. The linearized Langmuir isotherm Equation (7) is shown below:

$$C_e/q_e = C_e/q_{max} + 1/(K_L.q_{max}) \quad (7)$$

where C_e is the concentration of dye at equilibrium, q_e is the amount of dye adsorbed, q_{max} is the maximum adsorption capacity and K_L is the Langmuir constant related to the rate of adsorption. The parameter q_{max} can be calculated through the slope obtained from the plot of C_e/q_e versus C_e.

On the other hand, the Freundlich isotherm model proposes that the adsorption takes place at heterogeneous surface with uneven distribution of heat of adsorption over the surface. Unlike the Langmuir model, the Freundlich model takes into account multilayer adsorption. The linearized Freundlich isotherm equation (8) is given as,

$$\ln q_e = (1/n) \ln C_e + \ln K_F \quad (8)$$

where n is the empirical parameter, which is related to the strength of the adsorption process. The value of n falling within 1 to 10 indicates favorability of the adsorption process. The parameter K_F ($mg^{1-1/n}L^{1/n}g^{-1}$) is the Freundlich constant, and both K_F and n can be determined from the linear plot of ln q_e against ln C_e.

The Redlich-Peterson isotherm is another isotherm model that combines three parameters into an empirical isotherm. At high concentration, the Redlich-Peterson isotherm approaches the Freundlich model, while at low concentration, it follows the Langmuir equation. As this model incorporates three parameters, its equation can be applied to both homogenous and heterogeneous systems because of the high adaptability of its equation. The linear Redlich-Peterson isotherm equation (9) is given as,

$$\ln [K_R(C_e/q_e) - 1] = g \ln C_e + \ln a_R \quad (9)$$

where K_R and a_R are the Redlich-Peterson constants which can be determined from the linear plot of the above equation and g is the exponent which lies between 0 and 1.

All the three isotherm models gave reasonably high R^2 values, with the R^2 value of the Langmuir model being the highest at 0.9866. From the simulation plots as shown in Figure 2, despite the high R^2 (0.9462) of the Freundlich model, it is definitely not the correct fit for the adsorption of RhB onto TS. This can be further confirmed by the larger error values (Table 1) for this model. Although both the Langmuir and the Redlich-Peterson isotherm models produce similar simulation curves, based on both R^2 and overall error values, the Langmuir model is more suited to describe the adsorption process of RhB of TS. The size of the RhB molecule supports this experimental finding of forming a monolayer on the surface of TS.

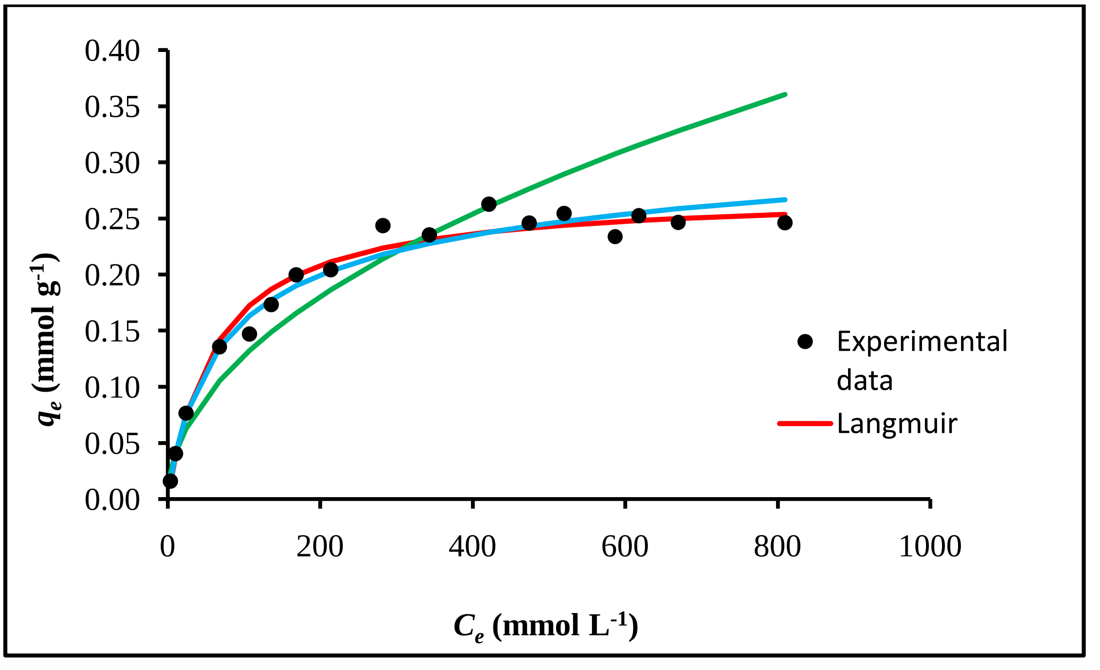

Figure 2: Simulation plots of the three isotherm models.

Table 1. Parameter values and error analyses for isotherm models used for adsorption studies of RhB.

Model	Values	ARE	SSE	HYBRID	χ^2
Langmuir					
q_{max} (mg g^{-1})	130.62	5.42	0.0047	0.14	0.01
K_L (L mmol^{-1})	0.02				
R^2	0.9866				
Freundlich					
K_F (mg$^{1-1/n}$L$^{1/n}$g^{-1})	6.22	14.90	0.0404	1.06	0.13
n	2.00				
R^2	0.9462				
Redlich-Peterson					
K_R (mmol g^{-1})	0.01	4.84	0.0050	0.14	0.02
g	0.90				
a_R (L mmol^{-1})	0.03				
R^2	0.9801				

The maximum adsorption capacity, q_{max}, based on the Langmuir model, of TS for the removal of RhB dye is 131 mg g^{-1} which is much higher than many reported adsorbents (Table 2), such as peat (85.5 mg g^{-1}), *Casuarina equisetifolia*needle (82.3 mg g^{-1}), kaolinite (46 mg g^{-1}) and cetyltrimethylammomium bromide modified sepiolite (26 mg g^{-1}). However, the q_{max}, of TS is lower when compared to modified adsorbents such as Fe_3O_4 nanoparticles with humic acid and polymer modified biomass whose q_{max} have been reported to be 162 mg g^{-1} and 268 mg g^{-1}, respectively. Nevertheless, as a natural biosorbent which is untreated and has not been chemically modified, TS can be considered as a very strong low-cost adsorbent for removal of RhB.

Table 2. Comparison of maximum adsorption capacity (q_{max}) of TS with other adsorbents reported for the removal of RhB dye.

Adsorbent	**q_{max}(mg g^{-1})**	**Reference**
Artocarpusodoratissimus (Tarap) skin	131.0	This work
Artocarpusheterophyllus (Jackfruit) seed	26.4	[26]
Jute stick powder	87.7	[36]
*Casuarina equisetifolia*needle	82.3	[37]
Fe_3O_4 nanoparticles with humic acid	162.0	[43]
Peat	85.5	[44]
Kaolinite	46.0	[45]
Cetyltrimethylammomium bromide modified sepiolite	26.0	[46]
Poly(methacrylic acid) modified biomass	268.0	[47]
Azollapinnata	72.2	[48]

Thermodynamics studies were conducted using an aqueous solution of 100 mg L^{-1} RhB at different temperatures ranging from 298 K to 343 K. The thermodynamics parameters, namely, standard Gibbs free energy change (ΔG^o), standard enthalpy change (ΔH^o) and standard entropy change (ΔS^o), for adsorption of RhB were calculated using standard thermodynamic equations as shown below:

$$\Delta G^\circ = -RT \ln K \quad (10)$$

and

$$K = C_s/C_e \quad (11)$$

where$K$ is the distribution coefficient for adsorption, C_S is the equilibrium dye concentration adsorbed (mg L^{-1}), C_e is the equilibrium dye concentration remaining in solution (mg L^{-1}), R is the universal gas constant (J mol^{-1} K^{-1}) and T is the absolute temperature (K).

$$\Delta G^\circ = \Delta H^\circ - T\Delta S^\circ \quad (12)$$

By combining Equation (12) and Equation (10), the following equation is obtained:

$$\ln K = (\Delta S^\circ/R) - (\Delta H^\circ/RT) \quad (13)$$

Thermodynamics studies showed that the amount of RhB dye adsorbed increased with increasing temperature, indicating an endothermic behaviour of adsorption. This is further supported by having a positive ΔH° of 11.32 kJ mol^{-1} for adsorption of RhB on TS. Further, negative ΔG° values are indicative of a spontaneous and feasible adsorption process, which is more favorable at higher temperatures. A positive ΔS°value of 46.31 J mol^{-1} K^{-1} suggests an increasing disorder and degree of freedom of the adsorption process.

3.3. *Regeneration study*

Regeneration studies were carried on TS to investigate its reusability and ability to regenerate the spent adsorbent. In this study, desorbing agents such as 1.0 M NaOH, 1.0 M HCl, distilled water and heating at 200 °C were used. Of these, heat treatment did not produce satisfactory results, whereby a significant reduction in the removal of RhB is observed as early as the first cycle. This can be attributed to the loss of organic functionalities at the treatment temperature of 200 °C, implying that the necessity of such functionalities for adsorption of RhB on TS. On the other hand, use of 1.0 M HCl, 1.0 M NaOH and simple washing with distilled water resulted in a reduction of 33%, 55% and 42%, respectively, after 4 consecutive cycles, while spent TS saw a reduction of 53% (Figure 3). Regeneration ability of TS with acid treatment is due to protonation of N atoms and the O atom in the cyclic ether component of RhB through which solubility is much improved, promoting desorption. Although the base treatment could ionize the carboxylic acid group, this process will not significantly improve solubility of RhB, and further, base treatment would destroy the surface features of TS. Treatment with distilled water would probably remove loosely bound (physisorbed) RhB molecules improving the adsorption ability of TS. Chemisorbed adsorbate species which are responsible for the formation of a monolayer, as per the Langmuir adsorption isotherm, would not probably be removed by distilled water treatment. Regeneration with HCl is thus recommended. However, there is a limit to the number of times the spent TS can be utilized.

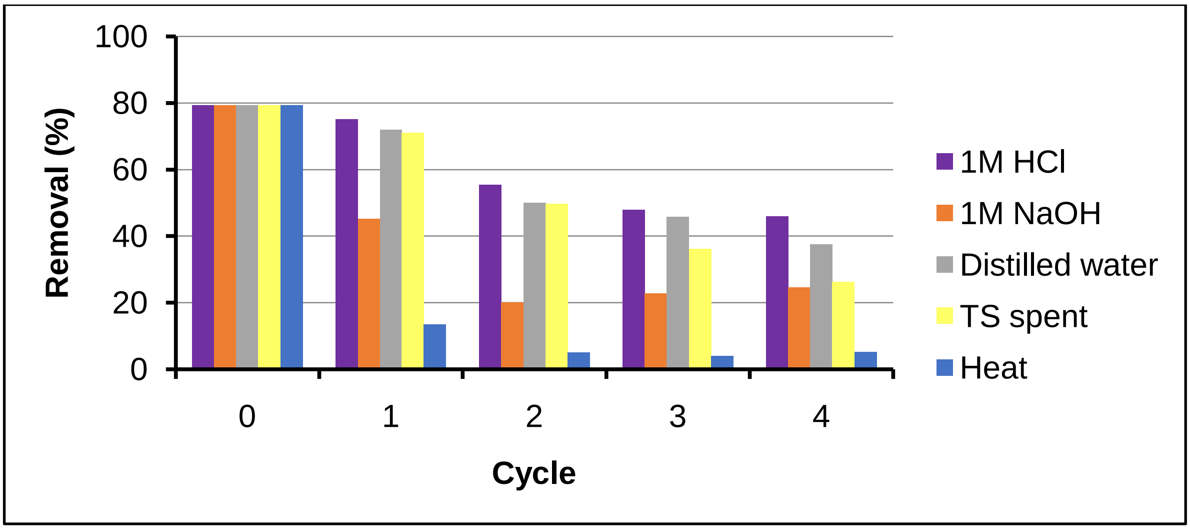

Figure 3.Cycles of regeneration of TS with RhB dye.

3.4. *Fourier Transform Infrared (FTIR) spectroscopy*

The FTIR spectra of TS, before and after treatment with RhB, are shown in Figure 4. Before adsorption, a broad peak was observed at 3402 cm^{-1} corresponding to the vibrations of -NH and -OH functional groups. The peak at 2920 cm^{-1} is characterized as a carbon-hydrogen containing species of attaching to alkane moieties, while peak at 1736 cm^{-1} indicates the presence of C=O group. Aromatic C=C functional group can be spotted as a sharp peak at 1620 cm^{-1}. The peak at 1061 cm^{-1} is due to C–O–C stretching and O-H bending.

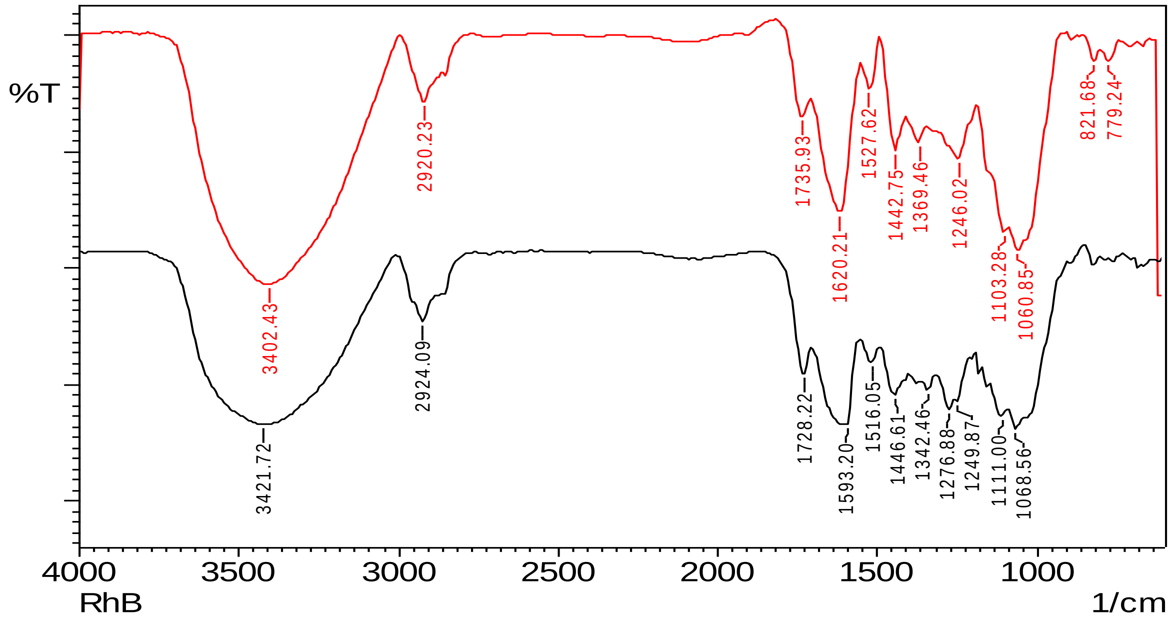

Figure 4. FTIR of TS before (red) and after (black) adsorption of RhB dye.

After adsorption of RhB dye, a visible shift was observed for many functional groups. The broad peak at 3402 cm^{-1} shifted to 3422 cm^{-1}. Both the alkyl CH group and the C=O group showed a slight downshift of frequency to 2924 cm^{-1} and 1728 cm^{-1}, respectively. The C-O-C stretching and O-H bending of lignin appeared at 1068 cm^{-1}. As RhB molecule has a conjugated system containing amine, ether and carboxylic acid functional groups, and as TS contains the above organic functionalities according to FTIR spectrum, many different types of intermolecular attractive forces, including hydrogen-bond formation, would be developed between the adsorbate and the adsorbent, demonstrating pH-independent removal of the RhB adsorbate. In addition, Coulombic attraction, whose strength depends on the medium pH, would also contribute to the removal of RhB by TS. Owing to many intermolecular attractions, it is proposed that the RhB molecule cover the TS surface with its flat orientation, favoring monolayer formation to support the Langmuir adsorption isotherm.

Conclusions

This study illustrates that TS can be successfully utilized as an adsorbent for the removal of RhB dye from aqueous solution. The adsorption process follows the Langmuir isotherm model with an attractive q_{max} value of of 131 mg g^{-1}, which is much higheras compared to many adsorbents reported. TS shows resilience and is not much affected by both medium pH and ionic strength, maintaining strong adsorption characteristics of RhB dye which makes it favorable in real wastewater treatment. Thermodynamics studies show that the adsorption process is endothermic and spontaneous. TS could be regenerated and reused maintaining satisfactory adsorption under HCl treatment condition even at the 4^{th} cycle. Based on the above stated desirable adsorption properties, TS has the potential to be used as a low-cost and effective adsorbent for the removal of RhB dye. The next logical step of this research would be to carry out adsorption studies with larger volumes to provide information for treatment of industrial effluents containing RhB dye.

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