Possibility of Utilizing the Lemon Peels in Removing of Red Reactive (RR) Dye from Simulated Aqueous Solution

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Abstract

Lemon peels (LP) were utilized as a biosorbent in separating the reactive red (RR) dye from simulated aqueous solution; several experiments were executed in this work to obtain a qualitative data to investigate the equilibrium and the kinetic in this paper. The effect of different parameters were investigated such as: the pH value, amount of LP, contact time, temperature, and initial dye concentrations to get their influence on the process of sorption. It is still crucial to investigate the complex fluid flow and mass transfer and the complicity of the interaction between adsorption and transport using low cost sustainable materials. The results of separation process in this work via adsorption process showed that the data of isotherm can be well characterized via Langmuir; this refers to that the reactive red dye be binding through two or more different sorption sites onto LP. The maximum RRdye uptake of 6.757mg/g was observed at pH2; according to

the Langmuir model; suggested that this peels can be categorized as an active sorbent in removing of RRdye from contaminated water sources. FTIR was recorded before and after sorption to show the position and number of the functional groups obtainable for RR binding on the sorbent which has been studied.

Keywords: Separation process, Fluid flow, Mass transfer, cost sustainable materials, Red Reactive (RR) Dye; Lemon peels; batch; Water treatment, adsorption; FTIR.

1 Introduction

Water is very important for the continuation of human beings and all creatures on earth. Industry of textile is one of the featured polluters' emissions of high concentrations of liquid waste through the environment. Above $7x10^5$ tones and about 10,000 specific kinds of dye and pigment are created throughout the world during a year; adding the volume is gradually increasing [1], [2]. Reactive-dyes are synthetic compoundshaving a pleasant and distinctive smell; that degrade with water and dispersed organic compounds. Dyes are the maximum without hardness identified contaminant due to their ability to seen by eye of human. While they are discharge to the water, they prevent the light of sun or any light from penetrating through also reduce water quality [1], [3].

Drainage of dyes may destroy the entire ecosystem and jeopardizes health of human [4]. Removing of dyes from liquids waste becomes important to the environment and it is needful not only to protect health of human but to protect the resources of water. Anywise, wastewater contaminated with dyes is complicated to treat [5]. Accordingly, there is a persistent need to develop and innovation processes; through which dyes can be efficiently removed. Process of biosorption which is utilizes inexpensive dead biomass to isolation of toxic pollutants. Biosorption is a well evaluated as a technique to remove the textile dyes from solutions [6]. There are many treatment processes such as adsorption and porous media were utilized to treat this type of containing effluent [7-12]. Activated carbon was used mostly as an adsorbent, because to high ability for sorption of organic matter; but its high cost so utilize of this sorbent is limited. For purpose to reducing the treatment cost; many attempts were made to obtain low cost substitution adsorbents. Many researchers were utilized some peels of fruit instead of activated carbon as it is expensive; therefore the use of natural residues from agricultural plants as adsorbent was reported to be attracted extensive attention for their presence in big amounts and at low cost; in addition to owing it that comparatively high particulate carbon content and being of porous structure [3], [7], [11]. The impact of sorbent builds on an essential range on their biochemical structure, in particular on functional collections that prepared in wall polysaccharidesof cell. Groups of carboxyl play an

active part in sorption of metal byalgal-sorbents [11]. Pectin, a cell-wall polysaccharide of higher-plants, is highly dependent on acid of galacturonic, it has a largenumberofgroups of carboxyl, and has a comprehensibility to cohesion together divalent cations. Hence, a material with rich pectin as citrus-peels, from which pectin is extracted commercially, has a high natural portability to binding metal. Many of rich pectin byproducts was evaluated due to their abilities of metal binding; which include: citrus peels, sugar beet pulp, apple waste, husks of coffee; Rice Husk, orange, banana peels, and variety of fruit materials, like a numerous types of citrus peel [10], [18]. In this work lemon peel was chosen as a biosorbent after drying at appropriate temperature. The commercial class of this dye with molecularformula ($C_{44}H_{24}O_{20}N_{14}C_{l2}Na_6S_6$), with molecular-weight of about (1496.98g/mol) and wave length (λmax) 540nm, and pH6.2, was obtained from [factory of Hilla-textile/Iraq (Origin: SIGMA-ALDRICH Company)]. Figure 1 below presents the structureof RRdye [19].
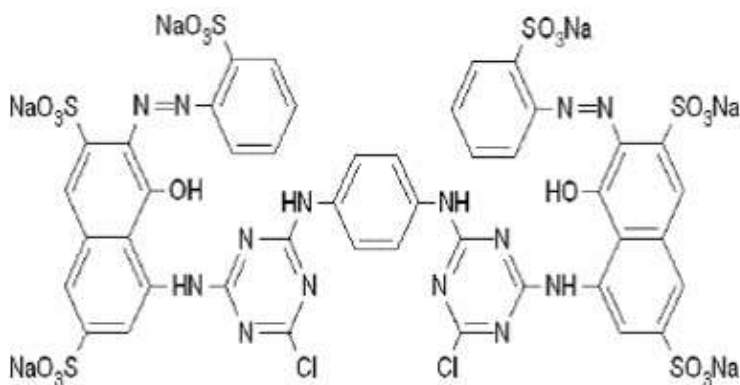

Fig. 1 The chemical structure of the RR dye formula

2 Experimental Study

In the current work, lemon peels (LP) were collected; cleaned with purified water. It dried at 105°C for 24 hr to remove moisture. It was grounded and sieved to 150 μm particle size; and storedin a closed botle for later use inadsorption studies, while in order to prepare a simulated RRdye solution by dissolving 1g of dry powder in 1000 ml of distilled-water and left in a glasss rectangular contaner of glazier at room temp.

The impact of: pH, LP, time; RRdye concentration were evaluated. Also the influence of temperature change on the biosorption capacity was assessed. All these experiments were executed in a batch process. The quantity removal of RRdye (q_e in mgg^{-1}) was assessed as [1]:

$$qe= \frac{V\,(Ci-Ce)}{M} \qquad (1)$$

Notice:

q_e : quantitative-ability-of-sorption (mg/g) (pollutant/adsorbent)

C_i :in itial concentratin of dye in solution

C_e : concentration of dye in solution after stability was reached

V : solution volume that is sorbent mass is visible

M: mass of sorbent [1].

All experiments were assessed aliquots of 250ml of RRdye solution about(50ppm) poured into conical flasks having exactly weighted quantityof LP extent from (0.25- 3)gm; in the batch experiments shaking the flasks at the time of equilibrium; then filtered by papers of filters in order to separate a suspended sorbent. Finally; RR concentrations were evaluated by UV-visible spectrophotometer. The pH of solution was detected by putting a proper ate quantity of hydrochloric acid and NaOH solutions (0.1N), pH was evaluated by using a pH meter.

3   Results and Discussion

3.1 Impact of pH

The pH impact on the sorption is caused by the fact of that vary of the pH value cause to variance in the properties of surface of the biosorbent and ionization degree [19]. In this work, capacity of bio sorption of RRdye by LP depending on the value of pH was assessed and the outcomesare represent in **Fig.2**; the capacity of biosorption was highly changed at acidic values of pH (from 2 to 5) and extend to the highest value of removal (about 77.4%; conditions:1hr, dose1gm, rpm200) at pH2. The LP surface has become a positive charge and this contributes to this sorption; this type of removing is property of anion sorption. The results calculated in this search are acceptance with those published in previous studies [20]. They concluded that more protons will be ready to protonated amine groups where they are in the acidic solutions to form groups, and that causing to increasing in sites of the biosorbent. It was found that at high pH the effective surface sites of the biosorbent may separated; so the capacity of biosorption was decreased. a high decrease in the capacity of biosorption was found when the pH is more than three and reached to the lowest value at pH value of seven [20]; this because to the reduction of capacity of sorption at these pH values is due to some effective groups on the LP surface which is more likely with less protonated or due to the competitiveness between OH⁻and dye anions [19-20]. So, the optimum pH value of 2 was utilized for the later experiments.
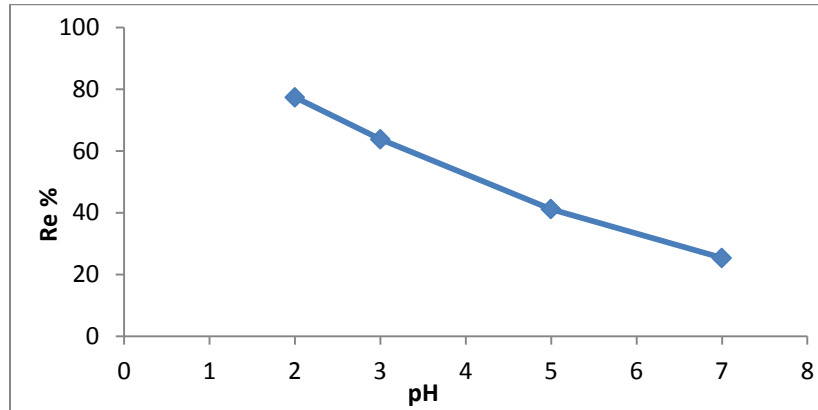
Fig. 2 Impact of pHvariation on removal rate

3.2 Impact of LP Quantity

The impact of adsorbent quantity was tested by varying the amount of PL from 0.5gm to 3.0gm in 250ml of simulated solution of 50ppmRRdye (Fig 3). After equilibrium the solutions were analyzed for the amount of dye. The results indicate that adsorption increased with increasing in adsorption quantity (from 1.5 to 3 gm), this assigned to an increase in the surface area of the absorbent and access to more effective absorption sites[21]; after this amount no change in removal efficiency was noticed because the adsorbent was sufficient to adsorb [21], [22]; so the optimum quantity that utilized in next experiments is 2.5gm.
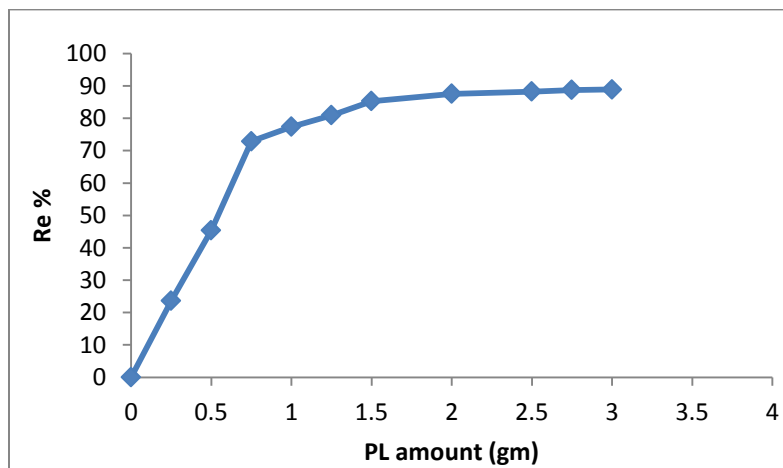
Fig. 3 Impact of change the quantity of PL on removal rate.

3.3 Impact of Contact Time

In Fig.4 contact time impact of LP on removing of RRdye was assessed at room temperature; the tests achieve at 180min. It is noticed that in the starting the rate of removal was higher till 120min (Re% is approximately 91%), and after that the rate of removal become practical very slows; and unchanged; because at the beginning all positions on the sorbent surface were empty and the solute concentration gradient was relatively high [9], [19]. Hence, the removal of dye minimize at increase in time of contact, which cause to that the numbering of empty positions on the dose surface [19], [20]. Because of that 120min was found to be the optimum time for the next tests.
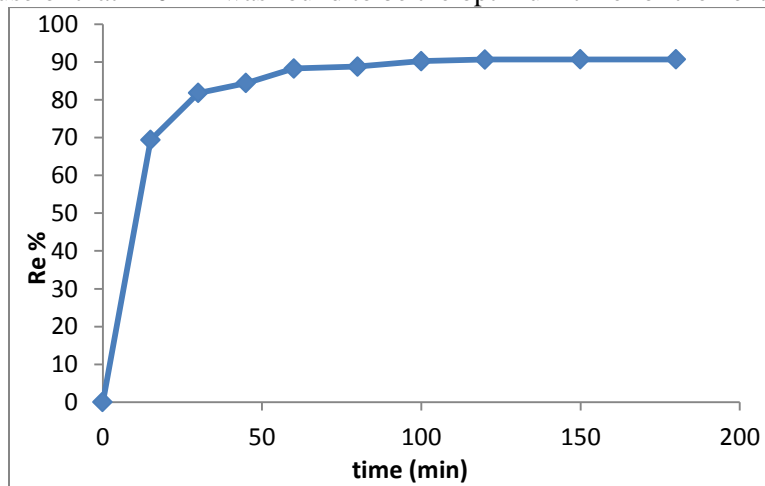

Fig. 4 Impact of contact of time on the removal rate

3.4 Impact RRDye Concentration

The varying concentrations of RRdyr (20-50ppm) in this work all the tests were rund at room temperature and mixing speed of about 200rpm. Fig.5 show clearly that increasing RRdye concentrations causes a decrease in the percentage of the removal rate at the exact adsorbent concentration (about 2.5gm of LP). This is due to the high initial concentration ratio of this molecules of dye to the available surface area; so sorption is affected by the starting concentration [9], [23].
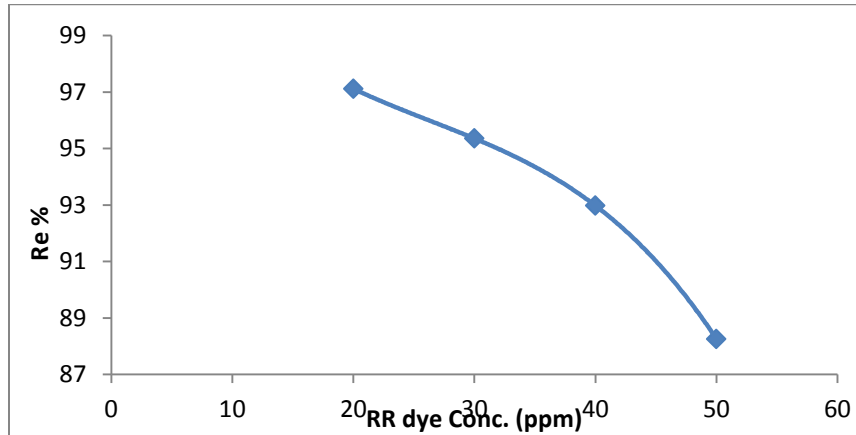
Fig. 5 Impact of change of RR Dye Concentration on removal rate

3.5 Isotherm and Kinetic Study

In this work, the experiential isotherm data of RR dye biosorption onto LP were modeled by two well-known sorption isotherm models which are Langmuir [24]:

$$q_e = \frac{q_m b C_e}{(1 + b C_e)} \qquad (2)$$

And Freundlich [25]:

$$\log q_e = \log K + \frac{1}{n} \log C_e \qquad \text{(Linear form)} \qquad (3)$$

The isotherm of Langmuir suppose monolayer sorption happened on a homogenous surface with a limited sorption sites of within the sorbent [24-26], while isotherm of Freundlich is appropriate to both monolayer (chemi-sorption) and multilayer sorption (physic-sorption) [**26**]. Both of Tabel1 and Fig 6 (a, b) showed the results of isotherm study. From this the data of isotherm can be well characterized by the Langmuir model.
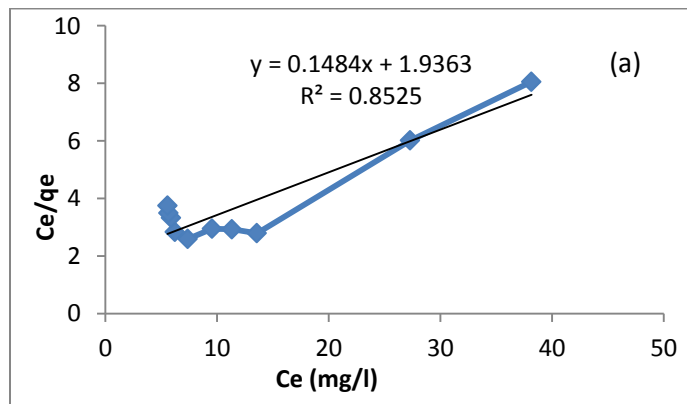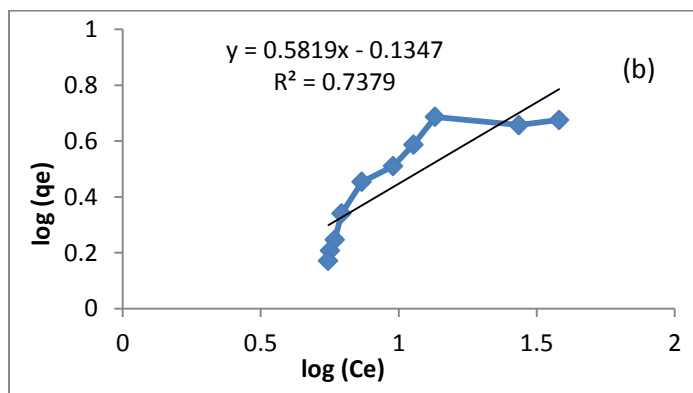
Fig. 6a.isotherms Parameters ofLangmuir model **b.** isotherms Parametersof Freundlich model

Table 1: Isotherms Parameters of both models

Langmuir model	RRdye	Freundlich model	RRdye
R^2	0.852	R^2	0.737
q_m(mg/g)	6.757	1/n	0.581
b (1/mg)	0.0764	K $(mg/g)(l/mg)^{1/n}$	0.735

Models of kinetic may be useful to understand the sorption mechanism, and estimating the sorbent performance. In the this search, the data of kinetic of sorption were resolved by pseudo-first-order, pseudo-second-order [27], [28]:

$$\ln(q_{eq} - q_t) = \ln q_e - k_1 t \qquad (4)$$

$$\frac{t}{q_t} = \left(\frac{1}{k_2 q_{eq}^2} + \frac{t}{q_{eq}}\right) \qquad (5)$$

Where q_{eq} and q_t (both in mgg^{-1}) are the quantity of dye adsorbed-at-equilibrium and at time corresponingly. K_1 (min^{-1}) and K_2 ($gmg^{-1}min^{-1}$) are the-rate-constants of kinetics for the models of pseudo first, and second-order [27].

The pseudo-first-order kinetic model has characterized sorption-in solid–liquid systems setup on the sorption ability of solids [27]; while the model of pseudo-second-order kinetic is instituted on the supposition that the sorption rate following second-order chemisorption [28]. The results are summarized in Table2 and Fig 7 (a, b).
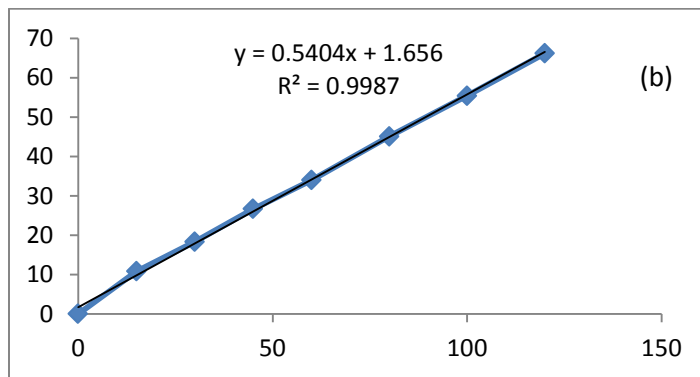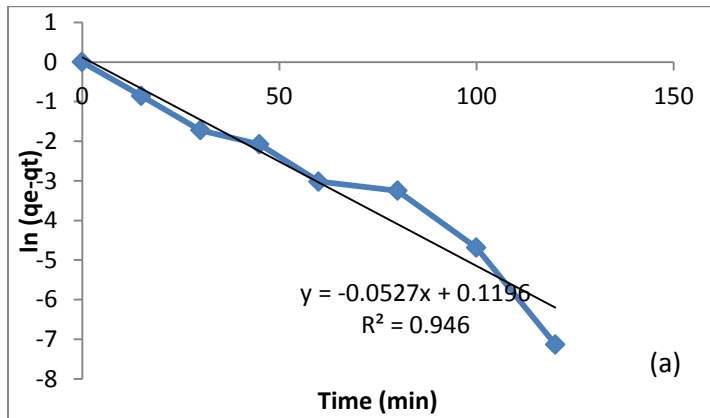
Fig.7 a. Parameters of Kinetic of pseudo first order model, andb. Parameters of Kinetic of pseudo second order model.

Table 2: parameters of both kinetic models

RRdye	q_e experimental mg/g	Pseudo-first-order			Pseudo-second-order		
		k_1 1/min	$q_{ecalculated}$ mg/g	R^2	k_2 g/mg.min	$q_{ecalculated}$ mg/g	R^2
	1.814	-0.052	1.126	0.946	0.176	1.852	0.998

### 3.6 Impact of Change in Temperature

RRdye adsorption on LP was assessed at temperatures of 298, 308, 318 and 328K (25, 35,45, and 55C°); shown in Fig.8. All parameters of thermodynamic which are: standard enthalpy ($\Delta H°$), standard Gibbs free

energy ($\Delta G°$), and standard entropy ($\Delta S°$), also the $\Delta H°$ and $\Delta S°$ values acquired from the next equations [23], [24]:

$$lnK_d = \left(\frac{\Delta S°}{R}\right) - \left(\frac{\Delta H°}{RT}\right) \tag{6}$$

$$\Delta G = \Delta H - \Delta ST \tag{7}$$

Notice: coefficient of distribution is K_d; T the enthalpy, and Temp in Kelvin; R is 8.314J/mol.K. The results of distribution coefficient and thermodynamic parameters are shown in Tables 3 and 4 below:
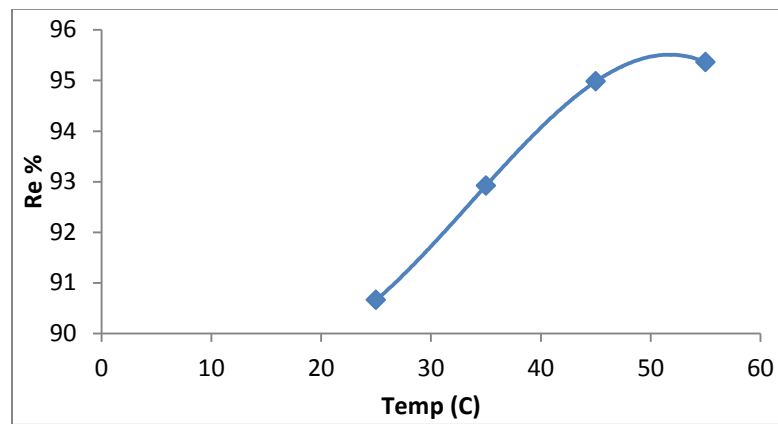
Fig.8 Influence of temperature change on removal rate

Table 3: The distribution coefficients at various temperatures which effect on removal rate

	k_d				R^2
RRdye	298 k	308 k	318 k	328 k	
	9.707	13.124	18.920	20.552	0.959

Table 4: The thermal parameters effects of RRdye adsorption on the LP

	ΔH(J/mol)	ΔS(J/mol K)	ΔG(kJ/mol)			
RRdye			298 k	308 k	318 k	328 k
	21.359	-90.778	-224.888	-232.456	-240.003	-247.551

It may be concluded that the removing rate of RRdye on LP was high at high temperature. Many authors recently worked towards green engineering and finding sustainable low-cost material on [29-35]. Recently, it was reported that the improve in sorption capacity when increasing temperature is back to excess mobility and spread of the ionic species [34]. An experience of adsorption can observe as a heterogeneous and inversion process at equilibrium [31] [34]. This pointing to the RRdye sorption is preferable to be at high Temp and endothermic, so that the adsorption range raised with T increasing.

3.7 FTIR Results

Analysis of FTIR was assessed to examin the functional groups on surface of LP (Fig.9). Table 5 shown all the band assignments. It can be seen from the current results of the FTIR spectra, the chemical formation of outcome particle of LP is related to the existence of minerals of carbonate inside the matrix of LP. On the other hand, to the resulting LP membrane molecules, the existence of amines and amides be expecting to have a positive charge functional groups like a: $–NH^+_3$ and $–CO–N^+H_2–$, depending on the pH of the solution [31].
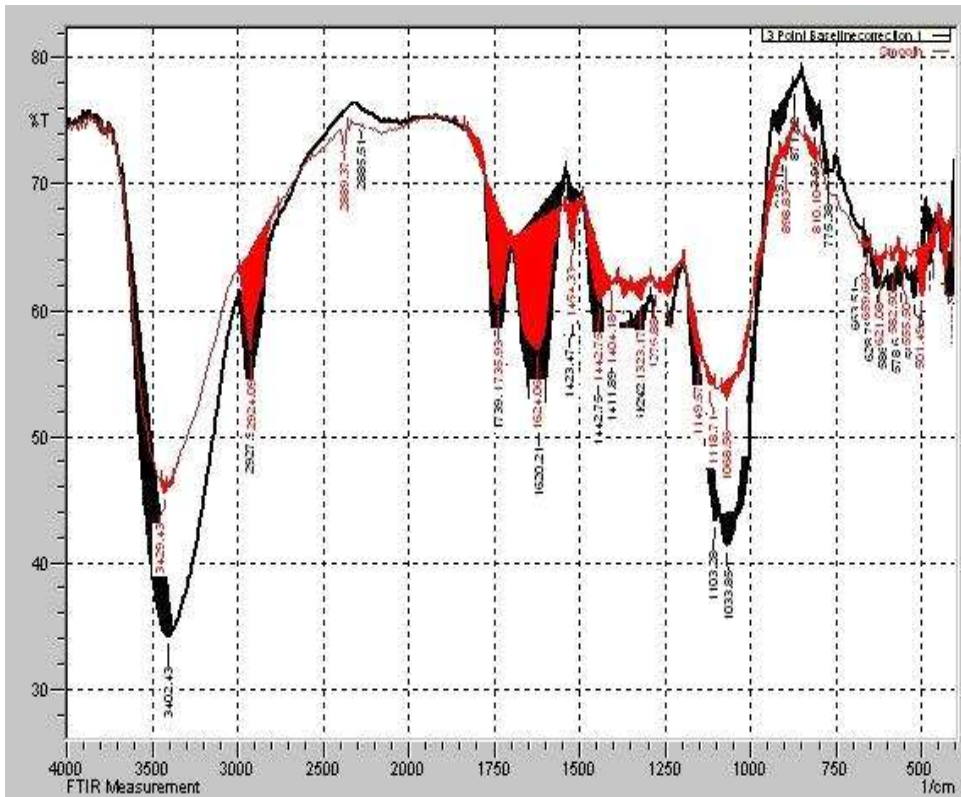

Fig.9 Analysis of FTIR of LP

Table 5. The band assignments (FTIR)

Waveform number (cm^{-1}) before adsorption of Cd(II)	Assignment Groups	Waveform number (cm^{-1}) after adsorption of Cd(II)
3402.43	Carboxylic acid, Amides, Alcohols, Amines	3429.43
2927.9	Alkanes, Carboxylic acid	2992.04
2885.51	Alkanes, Carboxylic acid	2889.37
1620.21	Alkanes, Alkenes, Carboxylic acid	1624.06
1423.47	Alkanes, Alkenes, Carboxylic acid	1454.33
1242.01	Alkanes, Alkenes, Carboxylic acid	1323.17
1103.28	Ethers, Ketones, Carboxylic acid	1118.21
1033.85	Ethers, Ketones, Carboxylic acid	1086.69
871.06	Alkenes, Anhydrides, Phosphines	898.83
775.38	Alkyl halides, Aromatic	810.10
628.11	Alkyl halides, Aromatic	659.66
518.6	Alkyl halides	582.50

4 Conclusions

This study assessed the chance of utilizing LP as sorbent in removing RRdye; this is a natural and variable material. The results appear that the LP can be as an active biosorbent in removal of RRdye from simulated solution and the maximum biosorption capacity was at pH2. The isotherm of sorption is modeled by the equation of Langmuir, so suggest the monolayer coverage ability of RRdye on LP surface. The model of pseudo-second-order kinetic exactly signalizes the kinetics of biosorption.

Acknowledgment

Reference

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