

## ISOTHERMAL STUDY OF CONGO RED DYE BIOSORPTIVE REMOVAL FROM WATER BY *SOLANUM TUBEROSUM* AND *PISUM SATIVUM* PEELS IN ECONOMICAL WAY

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**ABSTRACT.** *Solanum tuberosum* (STP) and *Pisum sativum* peels (PSP) were used for removal of toxic Congo Red dye using indigenous sources. Potato (*Solanum tuberosum*) and pea (*Pisum sativum*) are commonly and abundantly cultivated plants in Asian countries and their peels are easily available. The optimized conditions for *Solanum tuberosum* peels (STP) and *Pisum sativum* peels (PSP) were: adsorbent dose; 0.8 and 0.6 g, contact time; 50 and 35 min, pH; 3 and 2, agitation speed; 200 and 125 rpm and temperature; 30 °C for both STP and PSP respectively. Isothermal studies indicated that Langmuir model followed by equilibrium data more than Freundlich model. Negative value of thermodynamic parameter  $\Delta G^\circ$  shown that Congo Red removal by both adsorbents was spontaneous and exothermic in nature. The adsorption capacity ( $q_{max}$ ) for *Solanum tuberosum* and *Pisum sativum* peels were found to be 6.9 and 16.4 mg.g<sup>-1</sup>, respectively. Higher value of  $q_{max}$  for pea peels shown that it is more suitable adsorbent for removal of Congo Red dye than potato peels.

**KEY WORDS:** Congo red, *Solanum tuberosum*, *Pisum sativum*, Biosorption

### INTRODUCTION

Water is being polluted largely due to increased industrialization. Textile, paper and pulp, paint, printing and cosmetic industries use dyes. Discharge of waste water from such industries contains toxic organic solids. 50% of dye is lost during dyeing processes. The release of these dyes containing waste water into rivers, seas has adverse effects on the people who use such contaminated water for living purposes like washing, bathing and drinking. Even a small quantity of dye (1.0 mg/L) in water is detectable and objectionable. Besides this dyes can influence aquatic plants by reducing transmission of sunlight into water. Dyes are mutagenic, dangerous and cause harmful effects to humans [1]. Approximately 100,000 different dyes and pigments and over  $7 \times 10^3$  tonnes of dyes are manufactured annually throughout the world, about 10% of these might be present in waste water [2]. It is very difficult to treat waste water containing dyes because dye hinders light to penetrate into water, prevents photosynthesis and shows stability towards light and heat [3, 4].

Congo Red (CR) is an anionic, diazo, water soluble direct dye having maximum absorption at 520 nm [5]. It is mostly used in textile, paper, printing, leather and plastic industries [6]. It causes irritation to eyes and skin [7]. Different technologies such as: coagulation, chemical oxidation, filtration, membrane separation and biodegradation have been used for removing dyes from waste water but are not much effective. Adsorption is regarded to be an operative technique for waste water treatment owing to its ease, low price, simplicity and high effectiveness. Further, adsorption process can eradicate various types of contaminants therefore it has inclusive applicability in controlling water pollution. Several adsorbents have been reported in literature such as tea waste [8], saw dust [9], ackee apple seeds [10], spent mushroom [11], jute stick powder [12], bagasse fly ash [13], neem leaf [14], rice hull ash [15] and calcium-rich fly ash [16] for removal of Congo Red dye from aqueous solution.

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The cost and removal capacity of adsorbents is chiefly responsible for effectiveness of the adsorption. Currently, agricultural waste materials gain much more attention for the removal of dyes because of its low price and good effectiveness. *Solanum tuberosum* is commonly name known as potato. It is the world's fourth-largest food crop. It contains carbohydrates, vitamins and minerals and variety of phytochemicals [17, 18]. *Pisum sativum* is commonly name known as pea. It is a cool season crop in various parts of the world. It is involved in blood sugar regulation. It inhibits certain diseases like: arthritis and Alzheimer. It contains fiber, protein, vitamins, minerals and lutein [19]. The aim of this study was to check the usability of *Solanum tuberosum* peels and *Pisum sativum* peels as adsorbents for adsorption of Congo Red dye.

## EXPERIMENTAL

### *Reagents and instruments*

Congo Red dye ( $\lambda_{\max}$ : 520 nm), NaOH, HCl (Friends laboratory chemicals), Visible spectrophotometer 721 (China Yangzhou Wandong Medical Co., Ltd.), pH meter (HANNA™), Orbital shaker (model OSM-747) Digiteck instrument, Electric Grinder (National), Electric furnace (ranging from 30-1200 °C), Naberthem B170, FT-IR (Perkin Elmer-RXI with the range of 4000-700  $\text{cm}^{-1}$ ) and scanning electron microscope (JSM-6610) were used.

### *Preparation of adsorbent*

*Solanum tuberosum* and *Pisum sativum* peels were collected from domestic waste, washed with distilled water. They were kept in sunlight for drying and then ground in electric grinder to make fine powder. The same particle size of both adsorbents was made by passing ground powder through 60 mesh sieve (250 microns).

### *Preparation of adsorbate solutions*

A 0.1 g of Congo Red dye was dissolved in 100 mL of distilled water to make a stock solution of Congo Red dye. Standard solutions (5, 10, 15, 20, 25 mg/L) were made by taking (0.5, 1, 1.5, 2.0, 2.5 mL) of stock solution in 100 mL measuring flasks and diluted up to mark with distilled water.

### *Adsorption experiments*

Different factors on percentage adsorption were studied such as: contact time, adsorbent dose, temperature, pH and agitation speed. The dye solutions were filtered after biosorption experiment. Isothermal study was carried out by using Langmuir and Freundlich isotherm models. After adsorption, concentration of dye was calculated by measuring the absorbance of filtrate on visible spectrophotometer 721 after calibrating it with standard solutions of dye at 520 nm. Percentage adsorption and amount of dye adsorbed on STP and PSP was calculated by Eqs. (1) and (2), respectively:

$$\text{Adsorption (\%)} = (C_0 - C_e) / C_0 \times 100 \quad (1)$$

$$q = (C_0 - C_e)V/m \quad (2)$$

where q is the amount of dye adsorbed on biosorbent in mg/g.  $C_0$  (mg/L) is initial concentration and  $C_e$  (mg/L) is remaining concentration of dye which is determined spectrophotometrically, V is the initial volume of dye solution in litre and m is the mass of adsorbent in grams [20].

## RESULTS AND DISCUSSION

Characterization of adsorbents was done by taking FT-IR spectra (Figure 1) and by scanning electron microscope (Figure 2). The operational conditions were optimized separately for the selection of suitable biosorbent. Congo Red dye shows maximum absorption at 520 nm. All the absorbance measurements were made at 520 nm.

### Characterization of adsorbents

The results of physico-chemical properties of adsorbents are given in Table 1 and FT-IR spectra in Figure 1. FT-IR spectra of both adsorbents were compared to the literature for indicating the occurrence of various functional groups. FT-IR spectra study of *Solanum tuberosum* peels before adsorption showed band at 3341.74  $\text{cm}^{-1}$  which indicated the presence of O-H group while peak observed at 2923.77  $\text{cm}^{-1}$  showed the stretching of C-H bond. The peak at 1317.75  $\text{cm}^{-1}$  was assigned to the bending vibration of C-H group and band at 1148.72  $\text{cm}^{-1}$  indicated the stretching due to C-O group. The existence of these groups act as binding sites for adsorption. FT-IR spectra study of *Solanum tuberosum* peels after adsorption indicated that there was a change in wavenumber. It is observed from Figure 1 that some peaks were disappeared and new peaks were also appeared after adsorption of Congo Red dye on *Solanum tuberosum* peels. FT-IR spectra study of *Pisum sativum* peels before adsorption showed the following peaks of functional groups like 2919.69 and 2851.73  $\text{cm}^{-1}$  peaks for C-H stretching, 1625.62, 1618.61 and 1637.62  $\text{cm}^{-1}$  peaks for C=C stretching, 1317.75  $\text{cm}^{-1}$  peak for S=O group, 764.68  $\text{cm}^{-1}$  peak for aromatic C-H bending, 1100  $\text{cm}^{-1}$  and 1015.42  $\text{cm}^{-1}$  peaks for C-O stretching groups. The presence of these groups are responsible for adsorption of Congo Red dye molecule. FT-IR spectra study of *Pisum sativum* peels after adsorption indicated the similar peaks of functional groups but there was a slight change in wave number of some peaks like 1317.68  $\text{cm}^{-1}$  to 1318.81  $\text{cm}^{-1}$  and 1015.42  $\text{cm}^{-1}$  to 1009.60  $\text{cm}^{-1}$  of S=O and C-O functional groups, respectively. It is also observed from Figure 5 that some peaks were disappeared and new peaks were also appeared after adsorption of Congo Red dye on *Pisum sativum* peels [21, 22].

Table 1. Results for physicochemical properties of adsorbents.

Property	<i>Solanum tuberosum</i>	<i>Pisum sativum</i>	
pH	4.5	4.6	
Moisture content %	8	11	
Ash %	86	84	
Volatile organic compounds %	6	5	
Elemental analysis (mg/g)	Cd <sup>2+</sup>	0	0
	Ni <sup>2+</sup>	0	0
	Zn <sup>2+</sup>	0	0
	Pb <sup>2+</sup>	0	0
	Mg <sup>2+</sup>	0	0.0284
	Ca <sup>2+</sup>	0.021	0.036
	K <sup>+</sup>	6.26	1.96
	Na <sup>+</sup>	0.735	0.793
	Mn <sup>2+</sup>	0	0

SEM images of *Solanum tuberosum* and *Pisum sativum* peels are shown in Figure 2. They were indicating that dyes molecules had covered the rough surface of adsorbents, decreasing its roughness after adsorption process. From Table 1, it is clear that *Pisum sativum* peels had 84% ash, 11% moisture content, 5% volatile matter, while *Solanum tuberosum* peels contained 86% ash content, 8% moisture and 6% volatile matter. Low ash content of *Pisum sativum* peels shows small particle density so it acts as an effective adsorbent for biosorption. Higher value of

ash and volatile matter reduces the adsorption capacity of biosorbents. Results of elemental analysis indicated that toxic metals and micronutrients were absent while macronutrients were present in both biosorbents.

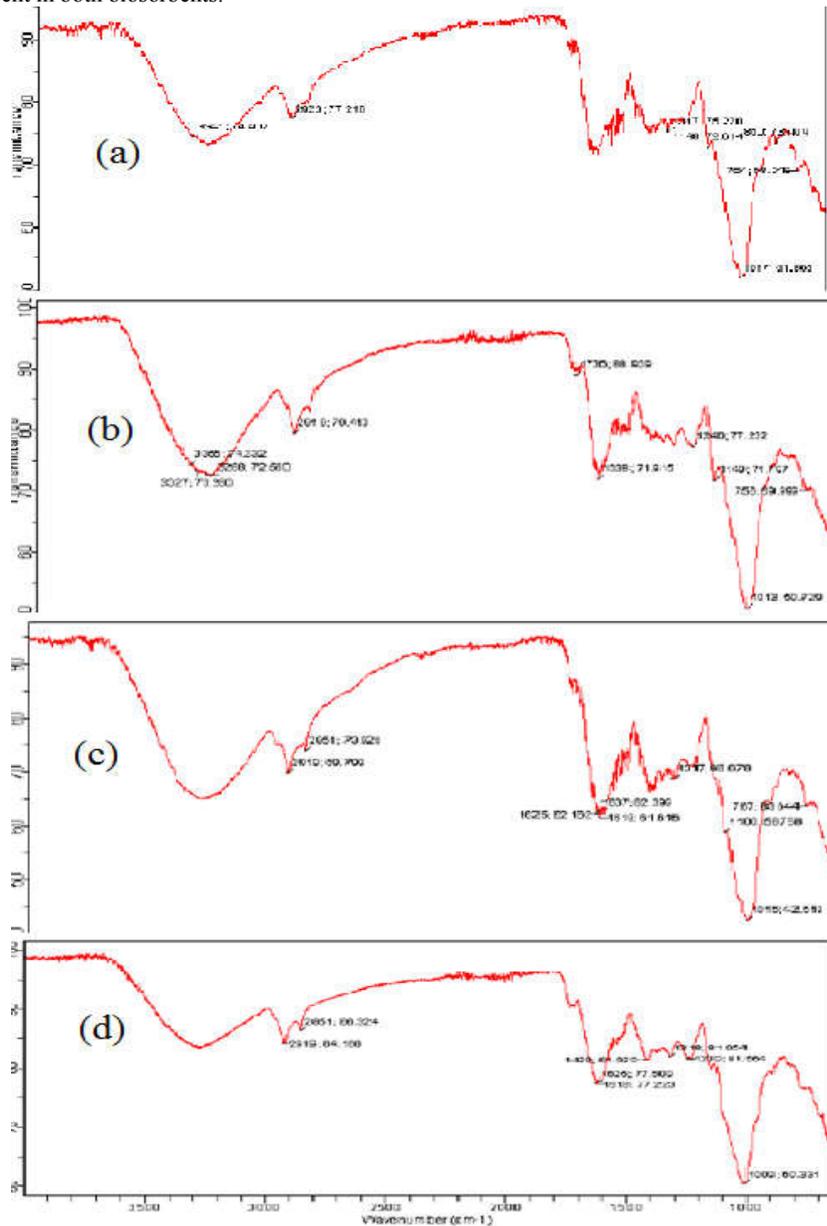


Figure 1. FT-IR spectra of *Solanum tuberosum* peels (a) before and (b) after adsorption, *Pisum sativum* peels (c) before and (d) after adsorption.

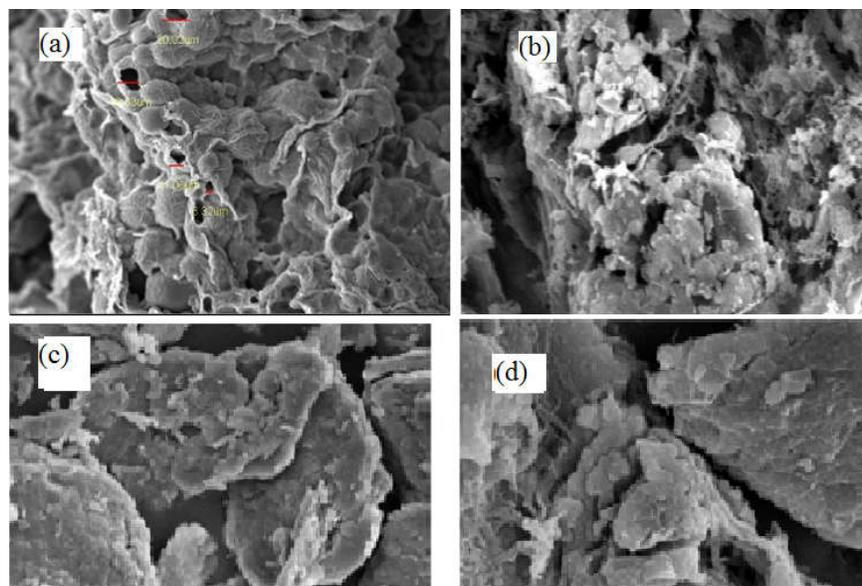


Figure 2. SEM image of *Solanum tuberosum* peels (a) before and (b) after adsorption, *Pisum sativum* peels (c) before and (d) after adsorption.

#### Determination of $pH_{pzc}$ of STP and PSP

To determine  $pH_{pzc}$ , experiments were carried out at different pH 2, 4, 6, 8 and 10 by adding 0.2 g of adsorbent to 50 mL of 0.1 M NaCl solution and agitated for 1 h in orbital shaker at 150 rpm and then allowed to settle for 48 h to reach equilibrium at 25 °C. The final pH was measured with the help of pH meter. A plot of  $\Delta pH$  versus initial pH determined the  $pH_{pzc}$  of *Solanum tuberosum* peels and *Pisum sativum* peels and it was 4.5 and 4.6, respectively. If pH of solution greater than  $pH_{pzc}$ , the adsorbent surface acquires negative charge and favors the uptake of cationic dye. If pH of solution less than  $pH_{pzc}$ , the adsorbent surface gets positive charge and adsorbs the anionic dye. Thus adsorption of Congo Red dye onto STP and PSP is favored at pH less than  $pH_{pzc}$  [23].

#### Effect of contact time

Figure 3 illustrates the result of comparative study of contact time by using *Solanum tuberosum* peels and *Pisum sativum* peels. It is evident that optimum contact time for *Solanum tuberosum* peels was found to be 50 min with percentage removal 39.9% while for *Pisum sativum* peels, it is 98.7% at 35 min. The trend of percentage adsorption of Congo Red was not regular. The reason behind the fact is that with increased contact time there is more time for dye molecules to make complex with the adsorbent [24].

#### Effect of adsorbent dose

Effect of adsorbent dosage on percentage removal of Congo Red was studied within the range of 0.2-2 g with a variation of 0.2 g. Graph illustrates comparative study of both adsorbents as a function of adsorbent dose. The percentage adsorption of Congo Red increased up to a certain extent then started decreasing. It was clear from Figure 4 that maximum percentage adsorption

of Congo Red 89.9% occurred at adsorbent dose 0.8 g for *Solanum tuberosum* peels and 96.1% at 0.6 g for *Pisum sativum* peels. By increasing adsorbent dose, percentage adsorption increases due to availability of more binding sites and surface area of adsorbent but percentage removal decreases with more increase in adsorbent dosage because adsorbent begins to aggregate and block the binding sites of adsorbent [25, 26].

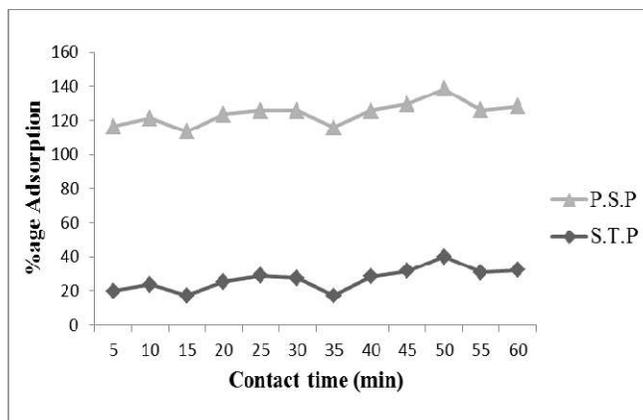


Figure 3. Comparative study of contact time on adsorption of Congo Red dye using *Solanum tuberosum* peels (STP) and *Pisum sativum* peels (PSP).

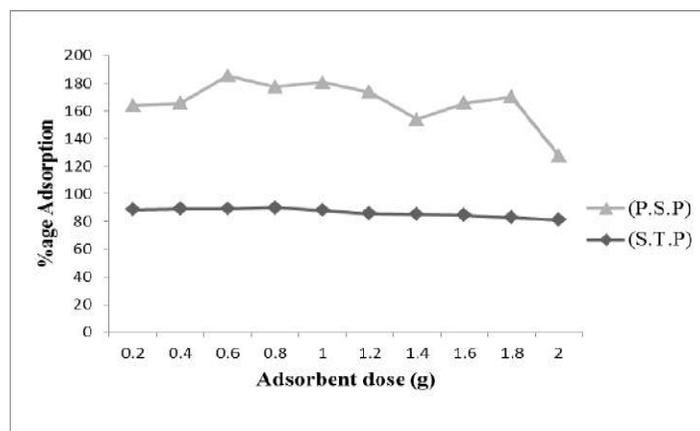


Figure 4. Comparative study of adsorbent dose on adsorption of Congo Red using *Solanum tuberosum* peels (STP) and *Pisum sativum* peels (PSP)

#### Effect of pH

From Figure 5, it is seen that optimized pH value for *Solanum tuberosum* peels and *Pisum sativum* peels are 3 and 2, respectively. *Solanum tuberosum* peels showed 97.9% and *Pisum sativum* peels showed 99.6% removal. The adsorption of Congo Red begins to decrease at higher pH. The reason behind is that, at higher pH, electrostatic repulsion occurs between negatively charged adsorbent surface and Congo Red dye (anionic type) while at lower pH,

concentration of  $H^+$  ion increases and adsorbent surface acquires positive charge. The strong forces of attraction occur between anionic dye molecule and the positively charged adsorbent surface at lower pH which causes maximum removal of dye [27]. Additionally, lower removal of anionic dye in alkaline condition is also by competition from excess hydroxyl ions for binding sites of adsorbent.

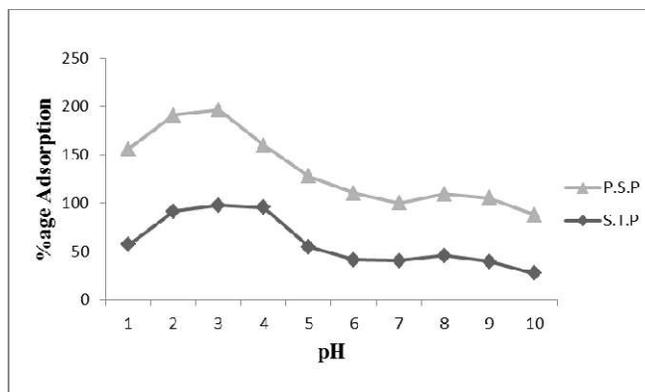


Figure 5. Comparative study of pH on adsorption of Congo Red using *Solanum tuberosum* peels (STP) and *Pisum sativum* peels (PSP)

#### Effect of temperature

The influence of temperature on adsorptive removal of Congo Red by both adsorbents was investigated from 20 °C to 70 °C. Figure 6 shows that maximum percentage removal of Congo Red dye was done at 30 °C for *Solanum tuberosum* peels with 43.6% adsorption. *Pisum sativum* peels showed maximum adsorption 94.4% at 30 °C. The percentage adsorption of Congo Red dye decreases with increase in temperature which shows that adsorption process is exothermic. The reason behind is that with increase in temperature, adsorptive forces between dye molecules and binding sites on the surface of adsorbent decreases [28].

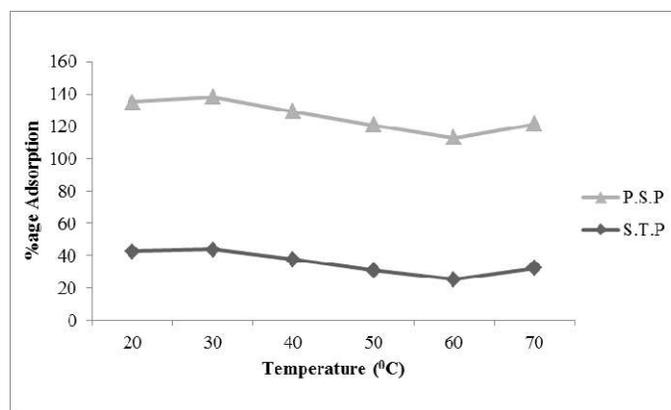


Figure 6. Comparative effect of temperature on adsorption of Congo Red using *Solanum tuberosum* peels (STP) and *Pisum sativum* peels (PSP).

*Effect of agitation speed*

Biosorption depending on agitation speed was studied, ranging from 25-200 rpm having an interval of 25 rpm each. From Figure 7, it is observed that maximum adsorption of Congo Red dye was occurred at 200 rpm by using *Solanum tuberosum* peels with 55.5% removal while *Pisum sativum* peels showed maximum removal of Congo Red dye at 125 rpm with 91.8% adsorption. The trend of percentage adsorption increases with increase in agitation speed. Shaking speed controls the biosorption process through film diffusion at lower shaking speed, thickness of fluid film around the particle of adsorbent is increased and rate of diffusion is decreased while at higher agitation speed fluid film becomes less thick and film diffusion is increased [29].

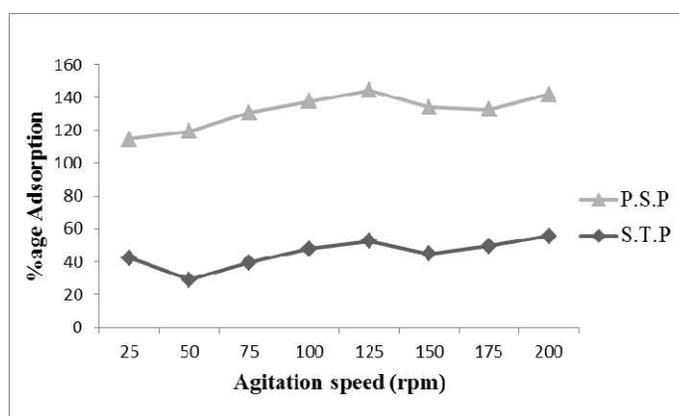


Figure 7. Comparative study of temperature on adsorption of Congo Red using *Solanum tuberosum* peels (STP) and *Pisum sativum* peels (PSP).

*Isothermal study*

Optimized conditions of each factor were applied for studying adsorption isotherm. Mechanism of adsorption was determined.

*Langmuir isotherm.* According to Langmuir adsorption isotherm, monolayer adsorption occurred. Langmuir isotherm equation is represented by following equation.

$$1/q = 1/(q_m b)C_e + 1/q_m \quad (3)$$

where  $q_m$  is monolayer (maximum) adsorption capacity (mg/g),  $C_e$  ( $\text{mg.L}^{-1}$ ) is the concentration of dye at equilibrium. When we plot a graph between  $(1/q_e)$  and  $(1/C_e)$ , a straight line is obtained with slope  $1/(bq_m)$  and intercept  $1/q_m \cdot b$  and  $q_m$  are Langmuir parameters. From the intercept and slope of plot, the value of  $q_m$  and  $b$  can be calculated.  $R_L$  is a separation factor. It describes the nature of adsorption process. It is represented by following equation.

$$R_L = 1/(1 + bC_0) \quad (4)$$

The value of  $R_L = 1$  indicates linear adsorption. In the present study the value of  $R_L$  is between 0-1. Comparative parameters of Langmuir isotherm for *Solanum tuberosum* and *Pisum sativum* is given in Table 2. The value of correlation coefficient ( $R^2$ ) for Langmuir isotherm of both

adsorbents indicated that Langmuir isotherm model followed by equilibrium data very well than Freundlich isotherm model. The higher value of  $q_{\max}$  (16.4 mg/g) for *Pisum sativum* peels shows that it is a good adsorbent for adsorption of Congo Red dye as compared to *Solanum tuberosum* peels [30, 31].

*Freundlich isotherm.* Freundlich isotherm is suitable to heterogeneous surfaces for adsorption and involves multilayer sorption. The Freundlich isotherm is represented as:

$$\log q_e = \log K_f + 1/n \log C_e \quad (5)$$

where,  $q_e$  (mg.g<sup>-1</sup>) is adsorption capacity of adsorbent at equilibrium,  $C_e$  (mg.L<sup>-1</sup>) is the equilibrium dye concentration. Adsorption capacity is described by  $K_f$  and  $n$  represents adsorption intensity. Both are Freundlich parameters. Their values are given in Table 2 and can be determined from the intercept and slope of the plot. The value of  $n$  for *Solanum tuberosum* and *Pisum sativum* was greater than 1 which showed favourable adsorption. It is clear from Table 2,  $K_f$  value of *Pisum sativum* was higher than *Solanum tuberosum* which indicated that *Pisum sativum* had more adsorption capacity for removal of Congo Red dye [31-33]

Table 2. Isothermal parameters for removal of Congo Red dye on *Solanum tuberosum* peels (STP) and *Pisum sativum* peels (PSP).

Langmuir isothermal parameters						
Adsorbent	Slope	Intercept	R <sup>2</sup>	q <sub>m</sub> (mg/g)	B (L/mg)	ΔG° (kJ/mol)
<i>Solanum tuberosum</i> peels	1.534	0.143	0.969	6.9	0.093	-5.8
<i>Pisum sativum</i> peels	1.337	0.061	0.901	16.4	0.045	-7.6
Freundlich isothermal parameters						
Adsorbent	Slope	Intercept	R <sup>2</sup>	K <sub>f</sub> (mg/g)(L/mg) <sup>1/n</sup>	n	
<i>Solanum tuberosum</i> peels	0.619	0.104	0.877	0.79	1.6	
<i>Pisum sativum</i> peels	0.777	0.066	0.862	0.86	1.3	

## CONCLUSION

In this study, peels of *Solanum tuberosum* and *Pisum sativum* were compared for adsorptive removal of Congo Red dye from water. *Pisum sativum* peels had more adsorption properties under optimized conditions for removal of Congo Red as compared to *Solanum tuberosum* peels. Thermodynamic study indicated that adsorption process was exothermic and spontaneous in nature. At low pH, adsorption of Congo Red occurred effectively. Equilibrium data was examined by using Langmuir and Freundlich isotherm. Results of isothermal study showed that Langmuir isotherm was well suited for removal of Congo Red. The higher value of  $q_{\max}$  for *Pisum sativum* peels suggests that it is good adsorbent as compared to *Solanum tuberosum* peels.

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