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Adsorption kinetics of iron (II) From waste/aqueous solution by using potato peel as carbonaceous material

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Abstract

Adsorption is one of the effective techniques for the removal of iron from aqueous solution / industrial waste water. The liberation of heavy metal pollutant particularly Fe (II) ions emerging out from industries has become a solemn concern. The objective of this study is to use low cost material as adsorbent for adsorption of Fe (II).The adsorption of Fe II from aqueous solution was studied using un-treated and treated (HCl& H_3PO_4) potato peel charcoal (PPC). Batch adsorption studies were carried out using different parameters like effect of contact time, effect of amount of adsorbent, effect of temperature and effect of concentration of adsorb ate. The test results revealed the adsorption capacity in following sequence: HCl- treated PPC > H_3PO_4-treated PPC > Untreated PPC. Adsorption studies were carried out by using Freundlich and Langmuir adsorption isotherms. Kinetic studies were revealed out by using pseudo 1st & 2nd order kinetics models to determine the rate constants. It was observed that the rate constants were generally decreased when concentration of solution was increased from 50 mg/L to 250 mg/L. It was also noticed that correlation coefficients (R^2) range from 0.912 to 0.983 for pseudo 1st order and 0.9956 to 0.9995 for pseudo 2nd order model.

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Introduction

Metal poisoning in water bodies poses serious environmental and ecological threats to the life on the planet earth. In recent past, metals like Zn, Co, Cu, Ni, Hg, Pb, Cr, Cd and Fe have produced significant toxicity in freshwater(Jong and Parry, 2004). Industries like rubber, electronics, electroplating, fertilizer, dyes, steel, paper, leather tannery and mining processes are the main sources of introduction of such metals in water (Selatnia *et al.*, 2004; Gundogdu *et al.*, 2009). Iron is regarded as secondary contaminant; It concentrates in various parts of the body including pancreas, liver, heart and responsible for diseases like hemochromatosis, cirrhosis and heart failure. Iron in the form of Fe II is soluble in water and is responsible for bad taste and green-reddish in color. Such water is unfit for the consumption and must be avoided.

Iron in the form of Fe III is insoluble and may cause drinking water to become turbid and iron precipitation [$Fe(OH)_3$] cause the blockage of water pipes and laundry staining which is difficult to remove (Bhattacharyya and Gupta, 2006; Tanwar *et al.*, 2008). The concentration of iron in drinking water must not exceed 0.3 mg/L(US-EPA, 2007). Researchers have adopted different methods to remove such contaminants from water including reduction, precipitation, flotation, adsorption, ion-exchange, filtration, electro-coagulation, lime softening and reverse osmosis (Shukla *et al.*, 2006; Chaturvedi and Dave, 2012). Removal of contaminants by adsorption using waste materials is desirable as such adsorbents are cheaper and economical in use.

Researchers have used different cost effective adsorbents including walnut shells (Wolfova *et al.*, 2013), citrus pectin (Balaria and Schiewer, 2008), and cotton waste(Riaz *et al.*, 2009) for Pb II removal, orange peel (Ajmal *et al.*, 2000) for Ni II removal, coconut shell charcoal(Babel and Kurniawan, 2004) and modified rice hull(Tang *et al.*, 2003) for Cr IV, Cashew nutshells (Tangjuank *et al.*, 2009) and Sugarcane bagasse (Ullah *et al.*, 2013) for Cr III, silica-gel(Zaporozhets *et al.*, 1998), modified coir fibers (Shukla *et al.*, 2006), bentonite clay (Tahir and Rauf, 2004), TBA treated kaolinite and montmorillonite (Bhattacharyya and Gupta, 2009) and chitosan(Ngah *et al.*, 2005) for Fe II ions and raphia palm (fruit endocarp)(Abasi *et al.*, 2011), hazelnut shells (Sheibani *et al.*, 2012), chitosan(Ngah *et al.*, 2005), ZrO-kaolinite, synthetic Na-A zeolite (Moaaz and Sridhar, 2016), libayan bentonite clay (Hamdi, 2017), commercialized chicken eggshells (Nasir *et al.*, 2016), and ZrO-montmorillonite (Bhattacharyya and Gupta, 2008) for FeIII ions from aqueous media. Potato peel waste was used for bio-reduction of perchlorate from water (Okeke and Frankenberger Jr., 2005), for treatment of pharmaceutical effluents(Kyzas and Deliyanni, 2015), Activated carbon from xanthoceras sorbifolia bange hull for the adsorption of Fe III, Co II and Ni II (Xiaotao *et al.*, 2017) and activated carbon/charcoal prepared from potato peel was used for removal of Cu II (Aman *et al.*, 2008; Moreno-Pirajana and Giraldo, 2011).

Nobody has reported potato peel charcoal for adsorption of Fe II to the best of our knowledge. The basic purpose of this research is the synthesis and use of environmental friendly low cost adsorbent. So we used potato peel charcoal for adsorption Fe II ions form aqueous solution.

Materials and methods

Reagents and materials

Analytical grade reagents were used for the entire research work. All the glassware used was acid washed and oven dried.

Adsorbent

Potatoes are abundant in our country so their peel is easily available as waste material. Potato peel was washed with deionized water, dried in oven and burned to charcoal at high temperature in a furnace (as reported in literature (Aman *et al.*, 2008). The resulting charcoal was ground to 0.2mm powder and stored for further use. Some of the Potato peel charcoal (PPC) was treated with H_3PO_4 and HCl separately. We studied the adsorption capacity of untreated-PPC, H_3PO_4–treated PPC and HCl-treated PPC on Fe II solutions.

Experimental procedure

Stock solution of 1000 mg/L was prepared by dissolving appropriate amount of $FeSO_4$. Stock solution was further diluted to prepare working standards of 50 mg/L and 250 mg/L Fe II solutions. Fresh working standards were prepared each time for calibration. All the experiments were carried out using 100mL flasks with 20mL of iron (II) solution and 0.1-1.0 g adsorbent (PPC) and stirred for optimum time at 150-200 rpm. The effect of temperature was studied in the range of 293–333 K. Stirred solutions were filtered using whatman no. 42 filter paper and Fe II concentration was determined by using Atomic Absorption Spectrophotometer (Unicam 969) operating with air acetylene flame, using hollow cathode lamp at 248.3 nm.

The percentage removal of Fe II was determined by using following equation(García-Rosales and Colín-Cruz, 2010).

$$R\% = \frac{C_o - C_e}{C_o} \times 100$$

and equilibrium adsorption capacity was determined by(Ramana *et al.*, 2012).

$$q_e = \frac{C_o - C_e}{m} \times V$$

Where q_e (mg/g) is the equilibrium adsorption capacity.C_o and C_e is the initial and equilibrium concentration (mg/L) of the Fe II ions in solution, V (liter) is the volume and m (g) is the weight of the adsorbent.

Adsorption Isotherms

Freundlich Adsorption Isotherm

Freundlich adsorption isotherm has been applied to study the adsorption process of 50 mg/L and 250 mg/L Fe II for HT-PPC.

The linear form of Freundlich equation (Ouadjenia-Marouf *et al.*, 2013) will be:

$$\log q_e = \log K_F + \frac{1}{n} C_e$$

Where q_e is the amount of metal adsorbed per unit mass of adsorbent at equilibrium (mg/g) and C_e is the equilibrium concentration (mg/L). K_F and n are Freundlich constants and K_F refers to the adsorption capacity (L/mg) of the adsorbent and index n is the heterogeneity factor.

Langmuir Adsorption Isotherm

The Langmuir equation in the linear form(Ouadjenia-Marouf *et al.*, 2013) will be:

$$\frac{C_e}{q_e} = \frac{C_e}{Q_o} + \frac{1}{K_L Q_o}$$

Where K_L and Q_o are Langmuir constants and describe energy of adsorption (L/mg) and maximum adsorption capacity (mg/g) respectively.

Following parameter indicates the isotherm shape(McKay *et al.*, 1980).

Kinetic studies

Lagergren pseudo 1st order and pseudo 2nd order kinetic models are used to determine the mechanism of adsorption of Fe II from aqueous medium.

Lagergren Pseudo First order model

The Lagergren kinetic model is the earliest known equation used to describe the adsorption process in liquid-phase systems.

The Lagergren pseudo 1st order equation can be expressed as(Tien and Ramarao, 2014).

$$\frac{dq_t}{dt} = k_1(q_e - q_t)$$

Where q_e and q_t are the amounts of metal adsorbed at equilibrium and at the given time t respectively. k_1 (min^{-1})is the pseudo first order adsorption rate constant. By integration, the linear form of above equation will be:

$$ln(q_e - q_t) = lnq_e - k_1 t$$

A plot of $ln(q_e - q_t)$ vs. t gives a straight-line with intercept of lnq_e. Value of k_1 can be determined from slope of the linear plot.

Pseudo 2nd order kinetic model:

Pseudo second order model can be described as(Gupta and Bhattacharyya, 2011).

$$\frac{dq_t}{dt} = k_2(q_e - q_t)^2$$

Where k_2 is second order rate constant. In linear form above equation will be

$$\frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e}$$

A plot of t/q_t vs. t gives a straight line with intercept of $1/k_2\ q^2_e$ and slope $1/\ q_e$. Value of q_e can be calculated and compared with that obtained from experiment.

Results and discussion

Sample characterization

The potato is a versatile carbohydrate rich food, highly popular world wide. The potato (*Solanum tuberosum*) is an herbaceous annual that grow up to 100cm tall. Its peel however has no use and therefore a cause of environmental degradation but potato peel charcoal has extensive surface area, which is a suitable attribute for Fe II adsorption.

The approximate analysis of properties of potato peel charcoal is shown in Table 1.

Table 1. Characteristic properties of potato peel charcoal.

Characteristics	Values
Ash (%)	7.01
Carbon (%)	27.85
Moisture (%)	1.5
Volatile (%)	60.85
Bulk Density (%)	0.52
Iodine number (mg/g)	725

Table 2. Values of Freundlich and Langmuir constants for Fe (II) adsorption on PPC.

Initial Fe II conc.	Freundlich Isotherm			Langmuir Isotherm		
	n	K_F (L/mg)	R^2	Q_o (mg/g)	K_L (L/mg)	R^2
50 mg/L	169.49	100.38	0.9225	24.57	9.09	1
250 mg/L	116.28	515.58	0.9699	121.95	1.038	0.9999

Effect of contact time on adsorption

20 ml of Fe II solution of initial concentration of 50 and 250 mg/L was stirred with 0.5 g of adsorbent at 293 K for a contact period ranging from 5–100 min separately. It is clear from Fig. 1 that percentage removal of Fe II, increased with the increase in stirring time and equilibrium was attained within 20 minutes for HCl-treated PPC and 40 minutes for untreated and H_3PO_4-treated PPC for all concentration ranges of Fe II. At any constant shaking time the percentage removal of Fe II decreased as the initial concentration of solution is increased. The percentage removal of Fe II decreased from 90.4% to 78% for HCl-treated PPC as the initial concentration of iron (Fe II) increased from 50 mg/L to 250 mg/L.

Effect of adsorbent concentration

Fig. 2 shows the percentage removal of Fe II at conc. of 50 mg/L and 250 mg/L. It is evident from the figures that the removal of Fe II increases with the increase of amount of adsorbent from 0.1 to 1 g. Best adsorbent concentration was found to be 0.8 g.

Table 3. Pseudo 1stand 2nd order parameters for Fe (II) adsorption on PPC.

Initial Conc.	Charcoal Type	Removal %	Pseudo first order kinetic model				Pseudo 2nd order kinetic model			
			q_e (exp.) (mg/g)	k_1 (min^{-1})	q_e (cal.) (mg/g)	R^2	q_e (exp.) (mg/g)	k_2 (min^{-1})	q_e (cal.) (mg/g)	R^2
50 mg/L Fe II concentration	Untreated PPC	36.1	0.44	0.112	0.515	0.912	0.44	0.5	0.4432	0.9956
	H_3PO_4 treated PPC	53.9	0.88	0.124	1.227	0.937	0.88	0.32288	0.8849	0.9971
	HCl treated PPC	90.4	1.808	0.103	1.5	0.983	1.808	0.206	1.81	0.9985
250 mg/L Fe II concentration	Untreated PPC	21.8	3.64	0.71	2.81	0.961	3.64	0.04437	3.649	0.9981
	H_3PO_4 treated PPC	44.3	5.4	0.69	4.267	0.983	5.4	0.03859	5.402	0.9977
	HCl treated PPC	78	7.8	0.136	8.864	0.972	7.8	0.0815	7.8125	0.9987

Effect of temperature

The effect of temperature, a major factor, influencing the adsorption was studied in the range of *293-333 K*. Removal efficiency was decreased with the increase of temperature. Decrease of removal efficiency might be attributed to increased desorption process with increase of temperature, but this decrease is not pronounced.

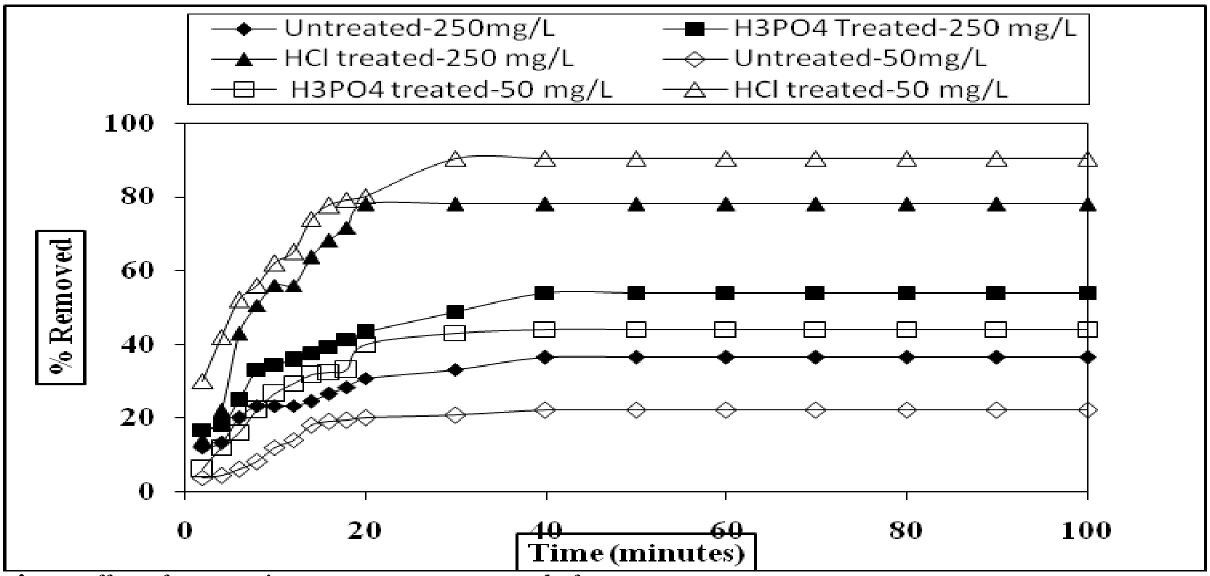

Fig. 1. Effect of contact time on percentage removal of Iron.

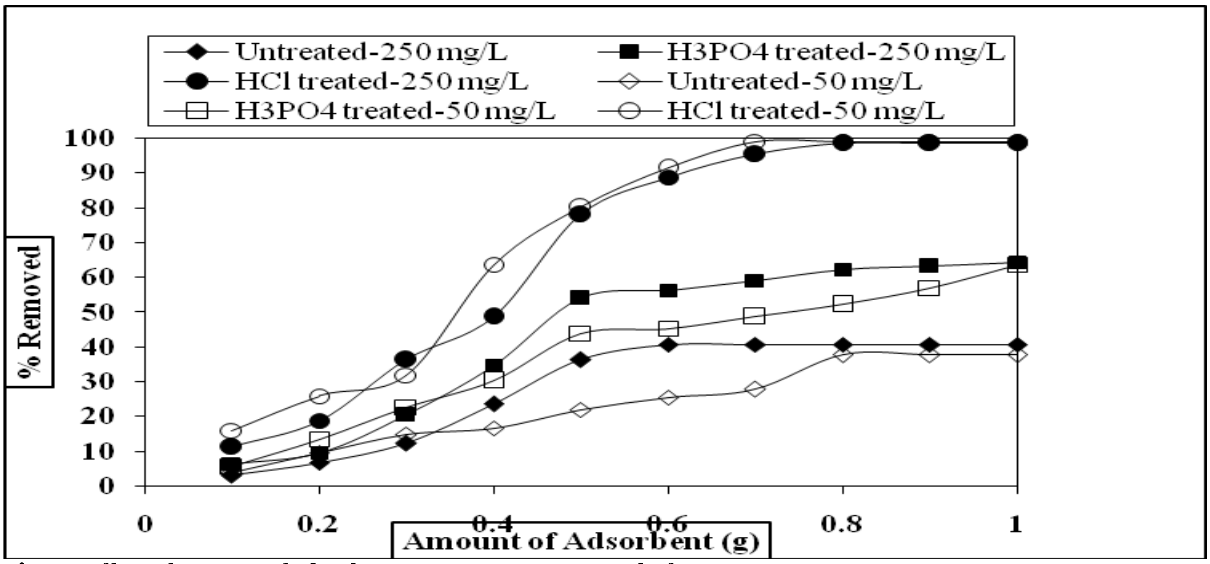

Fig. 2. Effect of amount of adsorbent on percentage removal of Iron.

This decrease in adsorption with increase of temperature indicates that the reaction is exothermic (Bekci *et al.*, 2006; Toor and Jin, 2012). Comparison of removal efficiency at different temperatures is shown in Fig. 3.

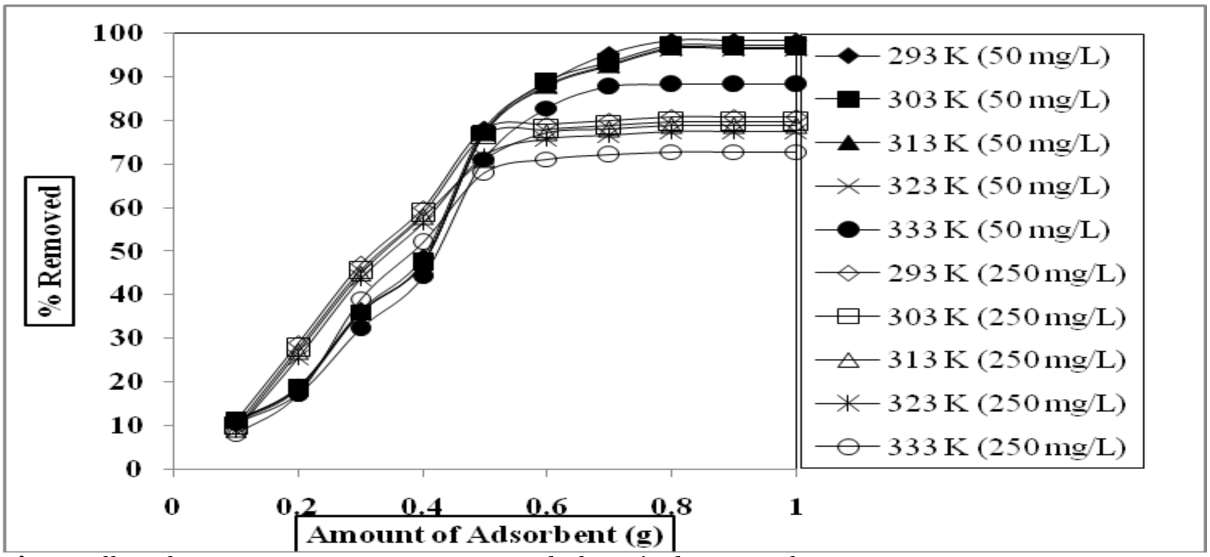

Fig. 3. Effect of temperature on percentage removal of Iron in the range of 293-333 K.

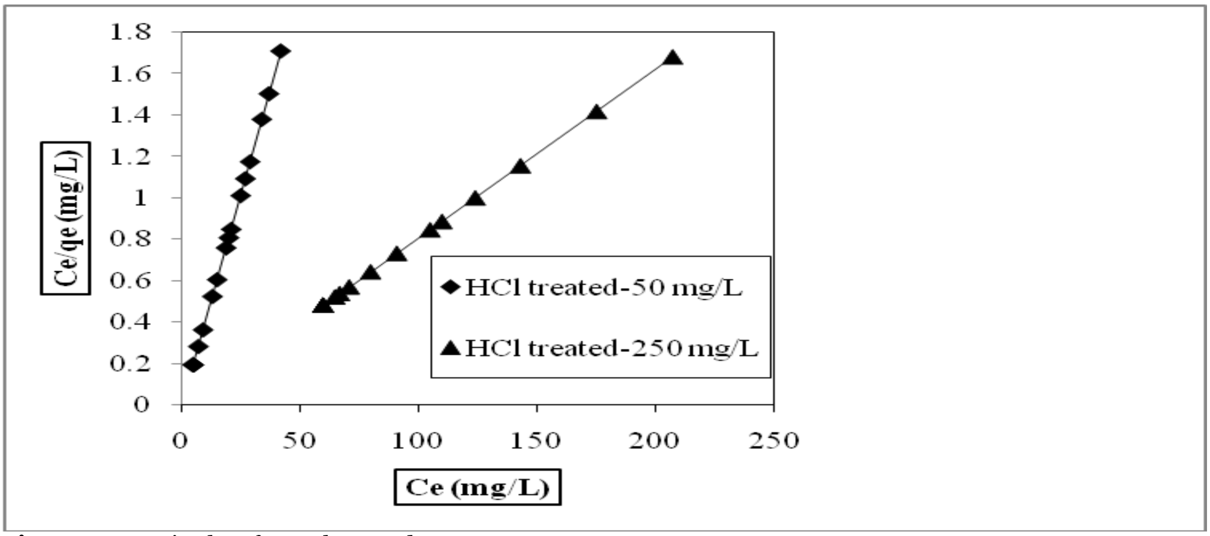

Fig. 4. Langmuir plots for HCl-treated PPC.

Adsorption Isotherms and Kinetic studies

Freundlich plot of *log* C_e *vs. log* $q_e$was drawn (for both 50 mg/L and 250 mg/L with HCl-treated PPC) and used to correlate with the experimental data, but it is assumed that adsorption process do not follow Freundlich isotherm as data is not fitted with linear regression (R^2 = 0.9225 and 0.9699) and bad linearity is achieved. Linear plots of C_e *vs.* C_e/q_e (for both 50 mg/L and 250 mg/L with HCl-treated PPC) showed that adsorption process seemed to follow Langmuir model (Fig. 4). Values of Freundlich and Langmuir constants are given in the Table 2. For Lagergren pseudo 1st order model, plots of *t vs. log* (q_e q_t) were drawn. Lagergren pseudo 1st order model do not explain the adsorption process as our calculated q_e value do not correlate with experimental q_e. Moreover, the correlation coefficient (R^2)is<0.99 which also indicates that the pseudo 1st order model is not able to explain adsorption mechanism therefore pseudo 1st order plot is also omitted. Plots of *t vs.* t/q_t were drawn for the correlation of pseudo 2nd order model and linearity of the plot and closeness of calculated and experimental q_e value showed that adsorption process appeared to follow pseudo 2nd order model. Pseudo 2nd order plots are given in Fig. 5. Table 3 shows values of q_e and rate constants *(k_1 and k_2)* of pseudo 1st and 2nd order model for all three types of adsorbents.

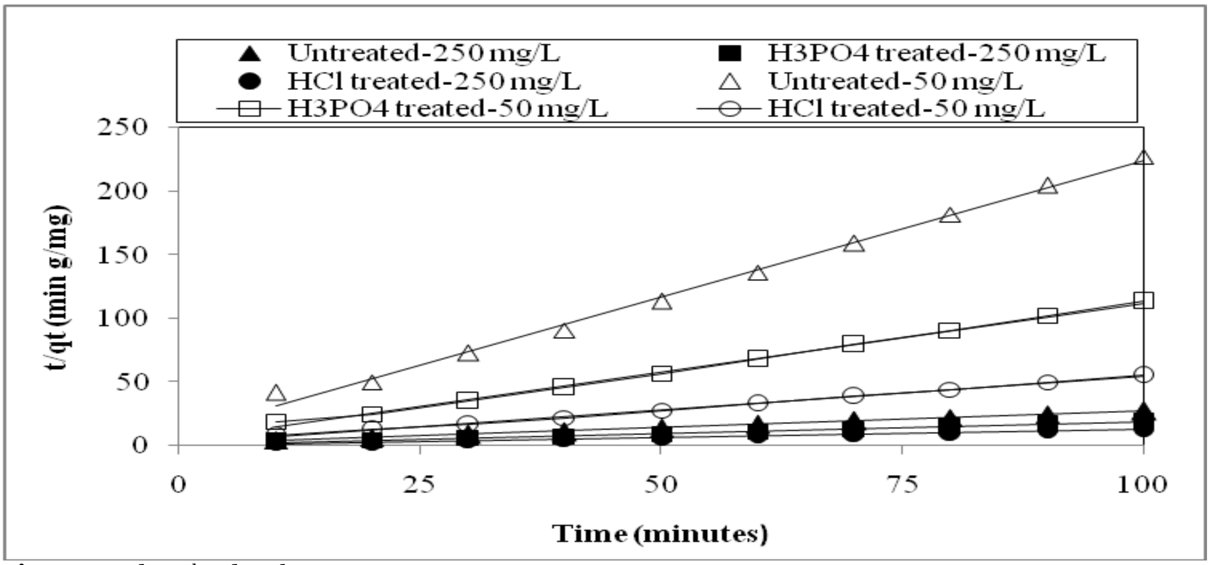

Fig. 5. Pseudo 2nd order plots.

Conclusion

From the above findings it is concluded that potato peels charcoal proved to be excellent adsorbent for the removal of Fe II from aqueous medium. 0.8 g PPC found to be enough to remove 90.4 percent of Fe II from an aqueous medium of 50 mg/L with a stirring time of 20 minutes.

The Fe II adsorption follows the Langmuir adsorption isotherm and pseudo 2nd order kinetic model. This study discovered that this new adsorbent is low-cost, local and easily available and has application for the removal of Fe II ions from aqueous medium.

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