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Single and Competitive Removal of Pb(II) in the Presence of Ni(II) using Polyacrylamide Grafted Rice Husk

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ABSTRACT: In a quest to find efficient adsorbents for metal ions, studies on various adsorbents for metals have been of interest since the past several decades. The present study is focused on the removal of Pb (II) ions from aqueous solution using poly acrylamide grafted rice husk by batch studies. Industrial waste waters generally contain metals present as mixtures, therefore the effect of Ni (II) on the removal of Pb(II) from mixtures of Pb(II)+Ni(II) ions has also been investigated. The adsorbent has been prepared by the treatment of rice husk with acrylamide in the presence of N,N-methylene bis acrylamide and potassium persulphate. The adsorbent has been characterised by infrared spectral studies. Maximum adsorption obtained is 93% at pH 5, metal ion concentration 300mg/L in 180mins at 298K. Isotherm analyses show that both Langmuir and Freundlich isotherm models are best obeyed. The process is endothermic and spontaneous in nature and follows pseudo first order kinetics. Intraparticle diffusion also occurs but is not the rate determining step. Application of Langmuir competitive model for the binary system shows that adsorption of Pb(II) has been supressed by presence of Ni(II) ions. Studies suggest that the adsorbent is effective and can find industrial applicability.

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Metals find widespread application in a large number of industries and thus are the major pollutants of the aquatic system due to the waste water discharge from these industries (Putra et al., 2014). Heavy metal pollution is a matter of concern, globally (Li et al., 2018; Medellin-castillo et al., 2017). The presence of heavy metal ions in surface water causes toxicity to aquatic flora and fauna and human life even at very low concentrations. Lead is the commonly found metal ions in effluents since a large number of industries (Khazaei et al., 2018; Bakkory et al., 2016). It is extremely toxic and tends to accumulate in bones, brain, kidney and muscles and also hinders the formation of haemoglobin in the body (Golkhah et al., 2017). The permissible limit for Pb(II) in waste water is 0.05-0.1mgL^{-1} and in drinking water the safe limit specified by various agencies is 0.010-0.015mgL^{-1} (Khazaei et al., 2018). It is therefore, essential to remove Pb(II) from wastewater before disposal. Numerous techniques such as chemical precipitation, coagulation and flocculation, membrane filtration, ion flotation, ion-exchange, electrochemical treatment and adsorption (Perumal et al. 2021) have been developed for the removal of metals but the adsorption process offers significant advantages due to simplicity of operation, high efficiency, easy metal recovery and possibility of regeneration of adsorbent in industrial applications (Medellin-castillo et al., 2017; Shih et al., 2019; Li et al., 2021). There are a large number of reports in literature on the use of agriculture-based

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residues and chemically modified agricultural based materials as adsorbents for the removal of metal ions (Medellin-castillo et al., 2017; Acharya et al., 2018; Sharma et al.2018). Reports reveal that chemically modified agricultural wastes are more effective for the remediation of heavy metal ions from waste water than unmodified agricultural wastes (Khalfa et al., 2016). Heavy metal removal via adsorption by polymeric adsorbents has been reported and is usually accomplished by the applications of polymeric ion-exchangers or by chelation (Nastasovic et al., 2004; Mohamed et al., 2010). A few reports are available on the use of hydrogels as adsorbents (Muya et al., 2016; Chowdhury et al., 2021; Shalla et al., 2019). In order to reduce the cost of the polymeric adsorbent, polyacrylamide coated agricultural wastes have been used as adsorbents. Polyacrylamide coated saw dust, rice husk (Raji and Anirudhan ,1998; Sharma et al., 2009); and polyacrylamide coated coconut husk (Sreedhar and Anirudhan, 2000) have been studied for the removal of Cd, Hg and Cr ions. Poly (acrylamide)-grafted coconut coir pith (Anirudhan et al., 2007) and poly(acrylamide)-grafted chitosan (Li et al., 2005) beads are known to act as efficient heavy metals adsorbents, Removal of cadmium from aqueous solutions by adsorption using poly(acrylamide) modified guar gum–silica nanocomposites has been studied by (Singh et al., 2009). With this in view the study of the adsorption potential and selectivity of polyacrylamide grafted rice husk for the removal of Pb(II) ions from aqueous solution using batch adsorption experiments has been undertaken. Since metals are generally not present as a single component in waste waters and few literature reports are available on the competitive removal of metals (Khalfa et al., 2016; Reed and Nonavinakere, 1992). The effect of the presence of Ni(II) on the adsorption of lead from binary metal system of Pb(II)+Ni(II) ions has been studied by Singh et al., 2020 using thiolated rice husk as adsorbent, Therefore, the objective of this paper is to investigate the single and competitive removal of Pb(II) in the presence of Ni(II) using polyacrylamide grafted rice husk from aqueous solution .

MATERIALS AND METHODS

Preparation of the Adsorbent: Rice (*Oryza sativa*) husk (variety PR 106) was procured from agricultural land of Sangrur, Punjab, India. It was washed thoroughly and dried in air, ground and sieved. 420µm sized particles were used for the present study.

Preparation of grafted rice husk The adsorbent can be prepared by the reported method (Raji and Anirudhan, 1996; Sharma et al., 2009), via graft polymerization process using N, N`-methelene bis(acrylamide) as a cross linking agent and potassium peroxydisulphate as an initiator. About 25 g of rice husk was taken and immersed in 300 mL of aqueous solution containing 2.5 g of N,N` - methelene bisacrylamide and 1.5 g of potassium peroxydisulphate . 7.5 g of acryl amide was added to this mixture and the contents were stirred vigorously at 70^0C. The polymerized product was washed with distilled water and dried at 80^0C. This polymerized mass was refluxed with ethylene diamine (25 mL) for 8 hours, washed with toluene and dried. To convert into cation exchanger, one part by weight of above material was refluxed with equal parts by weight of succinic anhydride at pH 4 for 6 hours, washed with 1, 4 –dioxane and finally with ethanol.

The reaction can be represented as:

$$R + CH_2 - CONH_2 + CH_2(NHCOCH_2)(NHOCCH_2)_2 \xrightarrow[70^0C]{K_2S_2O_8} RP - CONH_2$$

$$R + CH_2CH - CONH_2 + CH_2(NHCOCH_2)(NHCOCH_2)_2 \xrightarrow[70°C]{K_2S_2O_8} RP - CONH_2$$

$$RP - CONH_2 + NH_2CH_2CH_2NH_2 \xrightarrow[70°C]{K_2S_2O_8} RP - CONH_2$$

$$RP - CONHCH_2\,CH_2NH_2 + (CH_2CO)_2O \longrightarrow R\!-P\!-CONH(CH_2)_2NHCO(CH_2)_2COOH$$

Where: Rice husk is represented by R RP and denotes polymer coated rice husk

Preparation of Adsorbate Solution: Stock solution of concentration 300mg/L and working standard solutions (30-300mg/L) of Pb(II) ions were prepared in distilled water using $PbCl_2$ (Merck, A.R grade).

Binary solution for (Pb+Ni) system were obtained by varying the concentration ratios of Pb(II) + Ni(II) in the range 1:0, 1:1, 1:2, 1:3 and 1:4 for each fixed Pb(II) ion concentration of 30mgL^{-1}, 60mgL^{-1} and 90mgL^{-1} (Singh et al., 2018).

Characterisation of Adsorbent

Proximate analysis of rice husk: Proximate analysis of rice husk was carried out by heating 5g of the sample at 100°C for 1 hour to get the moisture content and further heating up to 700°C to determine the ash content.

Fourier Transform Infrared Spectroscopic (FTIR) analysis of polymer grafted rice husk: FTIR Spectra using Nicolet impact equipment with detector at resolution of 4 cm^{-1} were recorded as KBr pellets (i.e. mixture of 1mg powdered adsorbent with 100 mg KBr). The intensities of absorption bands were recorded for 32 scans per sample in the scanning range of 500 to 4000 cm^{-1}.

Batch Adsorption Studies: A known weight of adsorbent (250 mg) was added in 10 mL of metal ion solutions (300 mgL^{-1} to 30mg L^{-1} concentration) for regular time intervals varying from 30 to 210 minutes (till equilibrium was attained) at varying pH (2-8), adjusted by the addition of hydrochloric acid or ammonium chloride and ammonium hydroxide depending upon the pH desired. The resultant solutions were filtered and analyzed for metal ion concentration in single as well as binary metal solution. The metal concentration was determined titrimetrically (Singh et al., 2020; Vogel, 1978), using standard EDTA solution.

RESULTS AND DISCUSSION

Characterization of Adsorbent

Proximate analysis of rice husk: The moisture content at 100°C was found to be 5% and ash content at 700°C was 40.3%. The amount of silica in rice husk was determined gravimetrically and was found to be 12% (Abia and Asuquo, 2006).

The FTIR spectral analysis: The FTIR spectrum of the polymer coated adsorbent studied as KBr pellets show peaks at 3412 cm^{-1} (broad band) due to OH and NH group stretching, a peak at 2925 cm^{-1} attributed to CH_2 stretching, a band at 1522 cm^{-1} indicates NH group, C=O stretching is observed at 1669 cm^{-1} and another band at 1420 cm^{-1} due to C-O stretching is obtained, thereby showing the presence of COOH group (Sharma et al., 2009; Raji and Anirudhan, 1996).

Effect of initial metal ion concentration and contact time: Amount of metal ion adsorbed increases but percentage removal decreases with increase in initial metal ion concentration in the range 30-300 mgL^{-1} at 298K. This can be explained on the fact that at low concentration, the ratio of Pb(II) ions to the number of available adsorption site is low hence higher adsorption. Maximum removal takes place in the initial 30 minutes and equilibrium is attained at 180 min. which is due the decrease in the number of adsorption sites with time (Singh et al., 2018).

Effect of pH: The effect of pH has been studied in the pH range 2-8 (beyond pH 8 precipitation of Pb (II) ions occurs) (Medellin-castillo et al., 2017), for initial metal ion concentration 300 mg/L at contact time of 180 min. at 298K. The amount of Pb(II) adsorbed increases with pH from 2 to 5, further increase in pH shows a decline in amount adsorbed (Fig.1). At lower pH due to higher concentration of H^+ ions there is a competition with positively charged Pb(II) ions (Lawal et al., 2017; Cruz-Lopes,2021). With increase in pH the competitive effect of H^+ ions decreases and adsorption increases. Further increase in pH shows a decrease in the amount of Pb(II) adsorbed due to the formation of soluble or insoluble $Pb(OH)_2$.

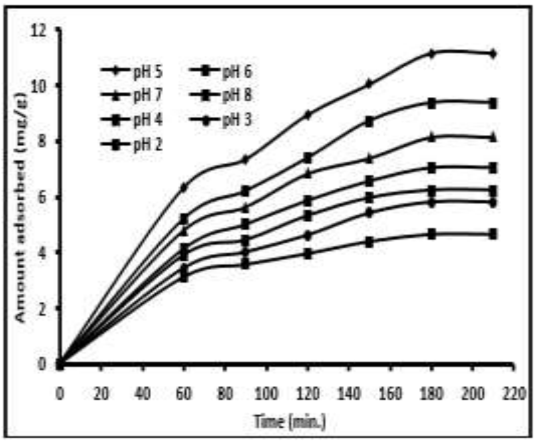

Fig 1. Amount of Pb(II) adsorbed on polyacrylamide grafted rice husk at different pH vs time at metal ion concentration of 300mg/L

Adsorption Isotherm Studies: The data obtained for adsorption at pH 5, and contact time of 180 minutes and metal concentration range 30-300 mgL^{-1} have been subjected to various adsorption isotherm models.

Freundlich isotherm: The Freundlich equation can be given as:

$$\log q_e = \log K_f + {}^{1}/_{n} \log C_e \qquad (1)$$

Where C_e is the equilibrium metal ion concentration (mgL^{-1}) and q_e is the amount adsorbed at equilibrium time (mg/g). The values of n and K, determined from slope and intercept of the straight line plot of log q_e vs. log C_e are given in Table 1. The value of 1<n<0 indicates the effectiveness of the adsorbent and high R^2 value of 0.991 suggests favourable and monolayer adsorption (Singh et al., 2018; Sharma et al., 2010).

Langmuir adsorption isotherm: Langmuir equation is given as:

$$C_e/q_e = 1/bQ_m + \left(1/Q_m\right) C_e \qquad (2)$$

Where C_e is the equilibrium metal ion concentration (mgL^{-1}), q_e is the amount of adsorbate adsorbed at equilibrium time for unit weight of adsorbent (mgg^{-1}), b is adsorption equilibrium constant related to apparent energy of adsorption (Lmg^{-1}) and Q_m maximum adsorption capacity (mg/g). The values of Q_m and b, as obtained from a plot of C_e/q_e against C_e, are given in Table 1. Results suggest homogenous monolayer adsorption on the surface (Singh et al., 2018) with maximum adsorption capacity of 7.50 mgg^{-1}.

A further analysis of Langmuir equation is made on the basis of R_L also known as separation factor,

$$R_L = 1/(1 + a_L\, C_o) \quad (3)$$

Where C_0 is the initial metal ion concentration (300 mg/L), R_L is separation factor. The values of R_L have been found to be 0.018 (between 0 and 1), suggests favorable adsorption (Singh et al., 2018).

Temkin isotherm: The linearised form of Temkin equation is:

$$q_e = B_T \ln A_T + B_T \ln C_e \quad (4)$$

Where, q_e is the equilibrium adsorption capacity (mg/g), B_T is a constant related to heat of adsorption (J/mol), T is absolute temperature (K) and R is universal gas constant, 8.314 J mol^{-1} K^{-1}. The values of the isotherm constants A_T and B_T obtained from the slope and intercept (Singh et al., 2018) of plot of q_e vs. ln C_e (Table 1), the value of R^2 value of 0.942 indicates that the Temkin isotherm fitted the data but the level of fitness was less than that of the Langmuir model and the Freundlich model.

Dubinin-Radushkevitch (D-R) isotherm: The expression for Dubinin-Radushkevitch isotherm is given as follows:

$$\ln q_e = \ln Q_m - K\, \varepsilon^2 \quad (5)$$

Where K ($mol^2\, kJ^{-2}$) is a constant related to adsorption energy, Q_m($mg\ g^{-1}$) is the maximum adsorption capacity, The polanyi potential Ɛ can be expressed as:

$$\varepsilon = RT \ln(1 + 1/C_e) \quad (6)$$

Where R is universal gas constant, 8.314 J $mol^{-1}K^{-1}$, T is absolute temperature (K) and C_e is the drug equilibrium concentration (mgL^{-1}). The values of K and Q_m are calculated (Singh et al., 2018) from the plot of ln q_e and ε^2. Adsorption parameters are given in Table 1. The value of correlation coefficient 0.963, suggests the applicability of D-R isotherm model. Using the value of K, the mean sorption energy E (kJ mol^{-1}) can be evaluated from,

$$E = K^{-1/2} \quad (7)$$

Value of E is 1.16 which lies b/w 1-16 kJ mol^{-1} indicating physical adsorption.

Error Analysis for Isotherm Studies: To optimise the best fit of isotherm equations to the experimental data, five different error functions of non-linear regression can be used for the evaluation of errors in isotherm curves.

$$SSE = \sum_{i=1}^{n} (q_{e,cal} - q_{e,exp})^2 \quad (8)$$

$$\text{(SAE)}: SAE = \sum_{i=1}^{n} \left| q_{e,cal} - q_{e,exp} \right|_i \quad (9)$$

$$\text{(ARE)}: ARE = \frac{100}{n} \sum_{i=1}^{n} \left| \frac{q_{e,cal} - q_{e,exp}}{q_{e,exp}} \right| \quad (10)$$

$$HYBRID = \sum_{i=1}^{n} \left[\frac{(q_{e,exp} - q_{e,cal})^2}{q_{e,exp}} \right]_i \quad (11)$$

$$\text{(MPDS)}: MPSD = \sum_{i=1}^{n} \left[\frac{q_{e,exp} - q_{e,cal}}{q_{e,exp}} \right]^2 \quad (12)$$

Table-1. Values of adsorption Isotherm constants for adsorption of Pb(II) on polyacrylamide grafted rice husk at pH 5 and 298K

Isotherm	Isotherm constants				Adsorption Capacity, (mg/g)	
				R^2	q_{exp}	q_{cal}
Freundlich	N 0.03	K_f (mg/g) 1.02		0.991	11.16	11.86
Langmuir	Q_m (mg/g) 7.50	B (L/mg) 1.80		0.952	11.16	11.17
Temkin	A_T (L/g) 1.80	B_T (J/mol) 3.19	b_T 776.6	0.942	11.16	11.45
D-R	K (mol^2/kJ^2) 0.37	Qm (mg/g) 22.4	E (kJ/mol) 1.16	0.963	11.16	14.17

Table-2. Error analysis values for the different isotherms for adsorption of Pb(II) on polyacrylamide grafted rice husk at pH 5 and 298K

Error function	Freundlich	Langmuir	Temkin	D-R
SSE	1.0×10^{-3}	0.991	1.26×10^{-3}	11.45
SAE	0.1×10^{-2}	0.45	0.25×10^{-2}	8.47
ARE	1.55×10^{-2}	0.23	0.17×10^{-3}	4.41
HYBRID	2.47×10^{-3}	1.04×10^{-2}	0.47×10^{-3}	0.99
MPSD	3.45×10^{-5}	2.42×10^{-3}	0.15×10^{-5}	0.74×10^{-2}

Adsorption Kinetics: A study of the adsorption kinetics has been carried out using different kinetic models.

Lagergren first order rate equation. Lagergren first order equation is represented by,

$$log\,(q_e - q) = log\,q_e - k_{ad}\ X\ t/\,2.303) \quad (13)$$

Where q_e and q (mg g^{-1}) are the amounts of Pb(II) adsorbed at equilibrium and at any time t (min) respectively, t (min) is the time of contact and k_{ad} is the adsorption rate constant (min^{-1}).

Pseudo second order rate equation: The data was further analyzed using pseudo second order rate equation.

$$t/_{q_t} = 1/_{K_2}\ X\ 1/_{q_e{}^2} + t/_{q_e} \quad (14)$$

Where K_2 is equilibrium rate constant for pseudo second order adsorption ($gmg^{-1}\ min^{-1}$), q_e and q_t (mg g^{-1}) is the amount of Pb(II) adsorbed at equilibrium and at any time and t (min) is the time of contact (Golkhah et al., 2017). A straight line plot for log(q_e - q) vs. t indicates the applicability this equation. Values of pseudo first order and second order kinetics parameters are given in Table 3.

Table -3. Kinetic parameters for adsorption of Pb(II) ions on polyacrylamide grafted rice husk at 298 K and pH-5

	Pseudo-first order			Pseudo-second order		
Pb^{+2}	K_{ad} (min^{-1})	q_e (mg/g)	R^2	q_e (mg/g)	K_2 ($gmg^{-1}\ min^{-1}$)	R^2
	1.88×10^{-2}	8.91	0.928	14.0	1.52×10^{-3}	0.975

Intra-Particle Diffusion Study: The possibility of intra-particle diffusion has been explored by Morris Weber equation:

$$q = K_p\ x\ t^{1/2} \quad (15)$$

Where q is the amount of drug adsorbed per unit weight of adsorbent at various time intervals (mgg^{-1}), K_p is the intraparticle diffusion constant ($mgg^{-1}\ min^{-1}$) and t is contact time (min.). K_p as calculated from the slope of the linear plot of q vs $t^{1/2}$ and has been found to be 0.73 $mgg^{-1}min^{-1}$. A straight line plot that does not pass through the origin is obtained. The two distinct regions due to external mass transfer effect indicate that both surface adsorption and intraparticle diffusion contribute towards the rate determining step (Singh et al., 2018). It can thus be inferred that the adsorption process follows pseudo first order kinetics and intraparticle diffusion contributes to the rate determining step.

Effect of temperature and thermodynamic study: Experiments conducted at temperatures 298K, 305K, 308K, 313K and 318K, for initial metal ion concentration 300 mg/L, at pH 5 for a contact time of 180 minutes with an adsorbent dose of 250 mg with particle size 212 microns, show that the amount of Pb(II) adsorbed increases with increase in temperature, probably due to increase in number of sorption sites generated because of breaking of some internal bonds near the active surface sites of adsorbent (Singh et al., 2018). The amount of Pb(II) adsorbed has been found to be 11.16 mgg^{-1} at 298K, 11.50mgg^{-1} at 305K, 11.78mgg^{-1} at 308K, 11.99 mgg^{-1} at 313K and 12.16 mgg^{-1} at 318K.

Thermodynamic parameters for adsorption of Pb (II) on polyacrylamide grafted rice husk: The thermodynamic parameters can be determined using equations:

$$\mathbf{ln}\,\boldsymbol{K_c} = \boldsymbol{\Delta S^o}/_{\boldsymbol{R}} - \boldsymbol{\Delta H^o}/_{\boldsymbol{RT}} \quad (16)$$

$$\Delta G^o = -RT\ln K_c \quad (17)$$

Where $K_C = C_{ad}/C_e$, is equilibrium constant and C_{ad} and C_e are the equilibrium concentrations (mgL^{-1}) of the metal ion on adsorbent and in the solution respectively **(**Golkhah et al., 2017). The values of ΔH^0 and ΔS^0 have been computed from the slope and intercept of the plot of lnKc vs 1/T and found to be 22.1 KJ mol^{-1} and 90.9 KJ mol^{-1} respectively, which indicates the endothermic nature of adsorption process and increase in randomness at the adsorbent–adsorbate interface during the adsorption, while negative value of -5.25 $KJmol^{-1}$ of ΔG^0 confirms the feasibility and spontaneous nature of the adsorption process (Singh et al., 2018).

Effect of mutual interference of Ni(II) ions on adsorption of Pb(II): Experiments were performed to investigate the influence of Ni(II) on the uptake of Pb(II) from binary solutions by varying the concentration ratios of Pb(II) + Ni(II) in the range 1:0, 1:1, 1:2, 1:3 and 1:4 for fixed Pb(II) ion concentration of 30mgL^{-1}, 60mgL^{-1} and 90mgL^{-1}. Percentage removal of Pb(II) ions decreases from 93.3 to 43.0%, 89.2 to 27.0% and 82.0 to 18.0% for increasing Ni(II) content at Pb(II) ions concentration of 30mg/L, 60mg/L and 90mg/L respectively (Fig. 2).

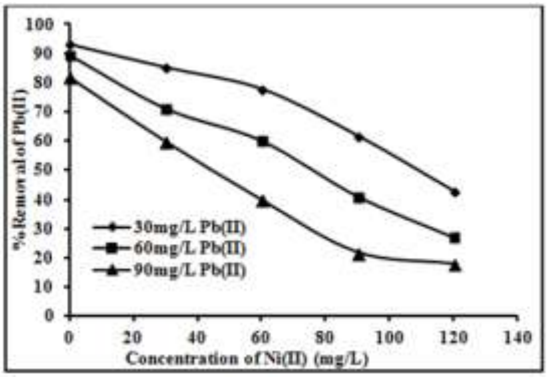

Figure 2. Percentage removal of Pb(II) from [Pb(II) +Ni(II)] mixture for various Pb(II) ion concentrations on polyacrylamide grafted rice husk at 298 K, at pH 5

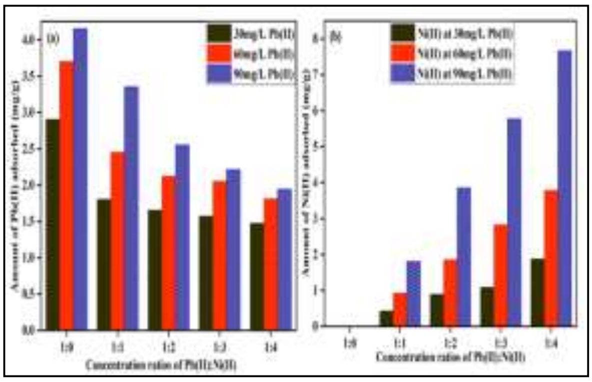

Figure 3. Adsorption capacity of (a) Pb(II) ions in [Pb(II)+Ni(II)] mixture (b) Ni(II) ions in [Pb(II)+Ni(II)] mixture at varying concentration ratios for contact time 120 min. at pH 5

The equilibrium data obtained from binary adsorption system have been analysed using q'_e/q_e ratios, where q_e is amount adsorbed for single component system at equilibrium and q'_e denotes the equilibrium amount adsorbed in the presence of other component (Singh et al., 2018). In general, three possible types of behaviour are exhibited: $q'_e/q_e > 1$ indicates Synergism (the effect of presence of the second component favours the adsorption of the primary adsorbate and adsorption from mixture is greater than that from the individual adsorbate solution), $q'_e/q_e < 1$ shows antagonism (the effect of the second component hinders the adsorption of the primary adsorbate and adsorption from mixture is less than that of the individual adsorbate in single component solution), $q'_e/q_e = 1$, non- interaction i.e there is no effect on the adsorption of the adsorbate in the presence of the other component in the mixture. The q'_e/q_e ratios are found to be 0.28 polyacrylamide coated rice husk, which indicates that the adsorption of the Pb(II) was suppressed by the presence of Ni(II) ions in the binary solution, hence the effect of the mixtures is antagonistic that is there is an inhibitory effect of one metal on binding of the other metal (Mahamadi and Nharingo, 2010).

The Langmuir competitive model (LCM): To further analyse the nature of competition among Pb-Ni ions, the Langmuir competitive model has been applied to the binary sorption equilibrium data using Eq. (18)

$$\left({}^{C_{e1}}/_{C_{e2}}\ X\ q_{e1}\right) = \left({}^{C_{e1}}/_{q_{m1}}\ X\ C_{e2}\right) + \left({}^{K_{L2}}/_{K_{L1}}\ X\ q_{m1}\right) \quad (18)$$

Where q_{m1} (q'_e), K_{L1} and K_{L2} are physical parameters, C_{e1} and C_{e2} are equilibrium concentrations of Pb(II) and Ni(II) in the mixtures of solutes. Values of (K_{L2}/K_{L1}) and q_{m1} (q'_e) of Langmuir competitive model were found to be $3.25x10^{-2}$ and 3.08 respectively. The value of $q_{m1}(q'_e) < 1$, indicates that the adsorption of Pb(II) was depressed by presence of Ni(II) ions in the binary solution (Tichaon and Jaco, 2013; Shoaib et al., 2011).

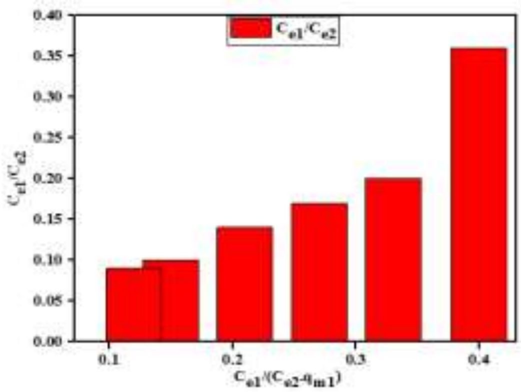

Fig 4. Langmuir competitive model for [Pb(II) (C_{e1}) + Ni(II) (C_{e2})], on polyacrylamide grafted rice husk for contact time 120 min. at pH 5, 298K

Conclusion: Results from equilibrium studies as well as isotherm and kinetic study analyses suggest that polyacrlamide grafted rice husk can be effectively used as an adsorbent for removal of Pb(II) ions from aqueous medium. Maximum removal occurs at pH 5 for a contact time of 180mins, the process is endothermic and follows pseudo first order kinetics and intraparticle diffusion contributes to the rate determining step. The study of binary mixture of Pb(II)+Ni(II) shows that the adsorption of Pb(II) has been suppressed by the presence of Ni(II) ions indicating that the effect of the mixtures is antagonistic in nature.

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