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Adsorption evaluation of selected heavy metal ions by amino-functionalized low-cost adsorbents. A Review

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ABSTRACT

Presence of heavy metals in drinking water has significant adverse effects on human wellbeing due to their toxicity nature. Several techniques have been employed to reduce their concentration to permissible levels. In recent years, adsorption has been widely investigated from low-cost adsorbents due to their cost effectiveness and easy in design. The application of amino-functionalized adsorbents for decontamination of wastewater have been reported. Generally, chemical modification of adsorbents has proved to have a higher adsorption capacity. Numerous amino-compounds such as ethylenediamine, 3-aminopropyl triethoxysilane, Triethylenetetramine, Sodium p-aminobenzoate, Acrylonitrile, Diethylenetriaminepentaacetic dianhydride, Polyaniline, Nitrilotriacetic acid, 3-aminopyrazole, N,N -dimethyl benzal aniline, Di (2-picolyl) amine, Ethylenediaminetetraacetic acid etc. grafted on adsorbents in studying equilibrium, kinetic and thermodynamics has been reported. These adsorbents are applied extensively in the removal of heavy metal ions such as Cu^{2+}, Cd^{2+}, Pb^{2+}, Ni^{2+}, Cr^{3+}, Co^{2+}, As^{5+} among others. The paper reviews the applicability of vast amino-functionalized adsorbents in the study of equilibrium, kinetics and thermodynamic adsorption studies of heavy metal ions from aqueous solutions.

Keywords: Adsorption, Heavy metals, kinetics, isotherms, thermodynamics, adsorption capacity

1. INTRODUCTION

Clean drinking water is vital for human and animal life sustainability[1]. However, intensive anthropogenic stress due to agricultural, industrial and technological activities has led to the release of contaminants to the natural water ecosystem[2]. Increased demand of water usage in agriculture, industries and at household levels has continued to increase loads of wastewater to the freshwater reserves making them unsuitable for consumption[3]. This has made safe drinking water scarce globally. Among the contaminants are heavy metals ions that are most toxic due to their non-biodegradability, persistence and carcinogenic nature once they accumulate for a long period of time[4]. They are discharged in aqueous chemical forms from storage battery manufacturing, alloy, metal plating, smelting, mining operations, radiator manufacturing, tanneries, radiator manufacturing and chloralkali industries amongst others [5, 6, 7, 8]. Also, natural activities of soil and rock erosion, weathering and rainwater via mineral dissolution, sorption/desorption of chemical agents and precipitation has also reported to release heavy metal ions to ground water bodies[9]. Among the metals, lead, copper, cadmium, zinc, nickel, mercury and arsenic are of global concern[10, 11]. When consumed and assimilated to the body tissues and organs such as kidney, bone, brain and muscles[12], they react with protein or enzyme ligands containing donor atoms of oxygen (OH, -COO,-OPO_3H, >C=O), sulphur (-SH, -S-S-) and nitrogen (-NH) [13] forming stable complexes altering the body's biochemistry and metabolism [14]. For example, they react with protein/enzyme sulphydryl groups (-SH and (-SCH_3) forming stable bonds[15] as shown in figure 1:

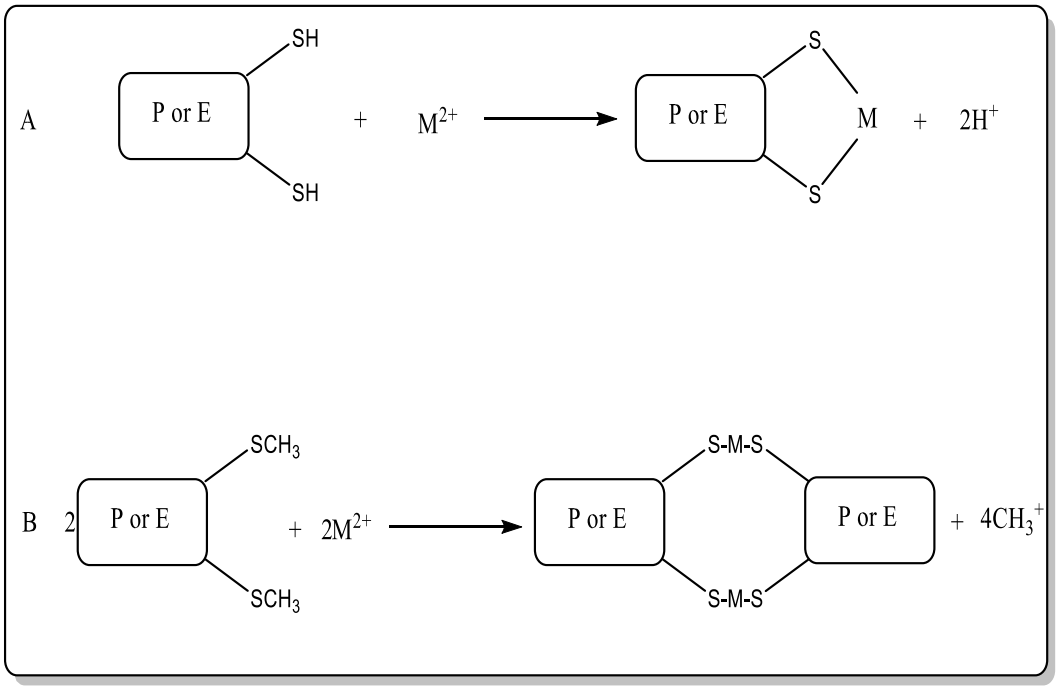

Fig-1: Biochemistry of toxicity
(A) Intramolecular bonding; (B) Intermolecular bonding; M = Metal; E = Enzyme; P = Protein[16]

Therefore, their prolonged exposure to human body leads to various acute and chronic health disorders such as hypertension, kidney damage, various cancers, skin irritations, mental retardations, dysfunction of the nervous system, anemia, dermatitis, rheumatoid arthritis and emphysema among others[17, 18, 19, 20, 21]. Therefore, treating heavy metal laden wastewater to permissible limits[22, 23, 24, 25] before discharge to the environment is paramount.

2. TECHNIQUES OF HEAVY METAL IONS DECONTAMINATION

Vast conventional techniques: ion exchange[18, 26, 27, 28, 29] electrodialysis[30], chemical precipitation[20, 31, 32, 33, 34], membrane filtration[35], coagulation/flocculation[36], flotation[37], adsorption[5] amongst others have been reported for their de-contamination. Among them, adsorption technique have increased research interest in recent years as the most effective method in water purification both at household and industrial level. This is due to its least expensive nature, availability of adsorbents from locally available materials in large quantities, easy in design and operation[6]. The discussion focused on adsorption.

3. ADSORPTION

Adsorption is a surface phenomenon where an adsorbate (metal ions) travels from metal solution and bind at the adsorbent surface by hydrogen bond interaction, electrostatic interaction and π-π interaction[38, 39, 40] . The technique has increased global popularity due to its simplicity in design and operation, economical, rapid, feasible, cost effective, easy regeneration and high removal efficiency[41]. The use of the adsorbents from agricultural wastes such as Jackfruit seeds[5.6] , mango peels[42, 43], cashew nut shell[44], orange peels[45, 46], avocado seeds[47], watermelon rinds[48, 49], cassava peels[50], chitosan[51] amongst others in heavy metals decontamination from wastewater have increased research interest due to their local availability in large amounts[52]. This has provided an alternative for an economically friendly alternative method of water purification. Researches have reported the use of the biomass wastes in their raw form to cause low adsorption capacity and poor selectivity towards metal ions, leaching of organic matter containing colouring agents and tannin compounds (secondary pollutants) in treated water increasing chemical, biological and total oxygen demands[6, 53, 54, 55] . Therefore, stabilizing the material via chemical modification have been reported to overcome above challenge. Studies have been reported on the chemical modification using amino compounds with the view of improving material stability in uptake of metal ions from wastewater.

4. ADSORPTION ISOTHERMS

They describe interactions of metal ions with binding sites, mechanisms of adsorption and adsorption capacity determination of the adsorbent at equilibrium[56]. They also determine equilibrium relationships between adsorbent sites and unadsorbed metal ions at a constant temperature[57]. The mathematical modelling of the isotherms are derived on assumption in relation to homogeneity or heterogeneity of the adsorbents, possibility of interaction between the metal ions species and the coverage type[58]. Isotherm models such as Koble-Corrigan (K-C), Temkin, Redlich- Peterson (R- P), Dubinin-Radushkevich (D-R), Sip's, Langmuir, Freundlich among others are used in isothermal analysis to describe concentration experimental data[5, 6, 59, 60, 61, 62, 63]. Freundlich and Langmuir models are most used models due to their simplicity[64].

4.1 Langmuir model

The model describes uptake of metal ions by adsorbent sites on a homogeneity monolayer coverage[5] with adsorbent sites having equal energy; chemically interaction of metal ions with fixed adsorbent sites and that each binding site hold one ion with no interaction between adsorbed metal ions[57, 65]. The monolayer coverage is assumed to remain constant even with higher metal ion concentration[58]. The linear form of Langmuir isotherm model can be presented by the equation:

$$\frac{C_e}{q_e} = \frac{C_e}{q_m} + \frac{1}{q_m b}$$

where q_e (mg/g) is the adsorbed metal ions at equilibrium, q_m (mg/g) is the maximum amount of metal ions adsorbed at equilibrium, C_e (mg/l) is the amount of the metal ions adsorbed at equilibrium. b (L/mg) is the Langmuir constant related to adsorption energy[66]. A straight line obtained from a plot of $\frac{C_e}{q_e}$ against C_e is used to obtain q_m and b from slope and intercept. 'b' values <1 are related to an increased affinity of the binding sites for metal ions adsorption[67].

Another characteristic of Langmuir separation factor (R_L) is used to define adsorption nature either unfavorable (R_L > 1), linear (R_L = 1), favourable (0 < R_L < 1) or irreversible R_L =0 [68].

$$R_L = \frac{1}{1 + K_L C_o}$$

4.2 Freundlich isotherm model

The model assumes a multi-layer coverage on a heterogeneous adsorbent surface with unequal adsorption energies[69]. The model is based on physical reversible and non-ideal interaction between the adsorbed metal ions in an adsorption process[65]. Freundlich linear equation can be expressed as:

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

where K_F and n correspond to Freundlich adsorption capacity (mg/g) constant and adsorption intensity constant respectively[70]. A linear graph obtained by plotting lnqe against lnce is used to determine K_F and n from intercept and slope respectively. The parameter ($\frac{1}{n}$) is related to adsorbent heterogeneity[71]. Smaller value of ($\frac{1}{n}$) < 1 indicates more heterogeneous adsorbent sites and close to 1 indicates more homogenous binding sites[72].

4.3 Sip's isotherm model

The model combines both Langmuir and Freundlich isotherm models[73]. Low concentrations assumes a multilayer coverage (Freundlich) and high concentrations assumes monolayer coverage (Langmuir)[74]. Its linearized expression is given by the equation:

$$\frac{1}{q_e} = \frac{1}{Q_{max} K_s} \left(\frac{1}{C_e}\right)^{1/n} + \frac{1}{Q_{max}}$$

where K_s (L/mg) and n are adsorption affinity constant and heterogeneity index and are obtained from a linear plot of $\frac{1}{q_e}$ against $\left(\frac{1}{C_e}\right)^{1/n}$ [75].

4.4 Dubinin-Radushkevich (D-R) isotherm model

The model describes the adsorbent porosity and adsorption free energy [76]. The model is usually applied to differentiate physisorption and chemisorption adsorption process of metal ions[77]. The model is expressed by equations:

$$q_e = Q_{DR} \exp\left(-K_{DR}\left[RTln\left(1+\frac{1}{C_e}\right)\right]^2\right),$$

$$\ln(q_e) = lnQ_{DR} - K_{DR}\left[RTln\left(1+\frac{1}{C_e}\right)\right]^2$$

Where q_e (mmol/g) is the quantity of adsorbed metal ions at equilibrium, Q_{DR} (mmol/g) is the maximum equilibrium adsorption capacity, K_{DR} (mol^2/ kJ^2) is the Dubinin-Radushkevich constant and C_e (mol/dm^3) is the amount of metal ions adsorbed at equilibrium[78].

The isotherm constant is related to the adsorption free energy, E (kJ/mol) which is expressed by the equation[76, 78]:

:

$$E = \frac{1}{\sqrt{2K_{DR}}}$$

Low values of E < 8 kJ/mol suggests a physisorption and > 8 kJ/mol suggest a chemisorption[78]. The table below shows a summarized adsorption isotherm data on different amino-functionalized adsorbents:

Adsorbent	Metal ion	Modifying Agent	Optimum parameters	Adsorption capacity	Adsorption isotherm model	Reference
Chitosan	Cu^{2+}	Acrylonitrile	pH= 5.0; contact time= 325 minutes; dose= 6 grams	230.79 mg/g	Langmuir	79
	Ni^{2+}		pH= 5.5; contact time= 300 minutes; dose= 5 grams	358.54 mg/g		
Chitosan	Cu^{2+}	Ethylenediamine	Contact time= 1 hour; temperature= 30^{o}C; adsorbent dose= 10 mg	32.30 mg/g	Langmuir	51
	Pb^{2+}			28.57 mg/g		
	Zn^{2+}			18.60 mg/g		
Jackfruit seeds	Cu^{2+}	Ethylenediamine	Dose = 10 mg; pH= 5.7; Agitation speed= 150 rpm; Time= 30 minutes; Initial concentration= 30 mg/L	32.97 mg/g	Langmuir	6
	Cd^{2+}		Dose = 10 mg; pH= 6.4; Time= 30 minutes; Agitation speed= 150 rpm; Initial concentration= 30 mg/L	24.75 mg/g		
	Pb^{2+}		Initial concentration= 30 mg/L; Time= 30 minutes; pH= 6.0; Agitation speed= 150 rpm; Dose = 15 mg	32.97 mg/g		
			adsorbent dose= 0.80 g; contact	35.71 mg/g		

Taro stalks	Cu^{2+}	Diethylenetriamine	time= 960 minutes; pH= 5.5-7.0		Langmuir	[80]
	Ni^{2+}		pH= 7.0-8.5; contact time= 360 minutes; adsorbent dose= 0.80 g	31.06 mg/g		
Micro-fibrillated cellulose (MFC)	Cu^{2+}	Aminopropyltriethoxysilane	contact time= 160 minutes and pH= 6.0	2.59 mmol/g	Sip's	[60]
	Cd^{2+}		contact time= < 8 minutes and pH= 6.0	1.45 mmol/g	Dubinin-Radushkevich	
	Ni^{2+}		contact time= 35 minutes and pH= 6.0	1.05 mmol/g	Dubinin-Radushkevich	
Graphene oxide hydrogel	Pb^{2+}	Polyethylenimine	pH= 7.0; time= 6 hours; temperature= 333 K	602 mg/g	Langmuir	[81]
	Hg^{2+}			374 mg/g		
	Cd^{2+}			181 mg/g		
Pineapple leaf fiber	Pb^{2+}	Ethylenediaminetetraacetic acid	pH= 6.0; time= 90 minutes	63.92 mg/g	Langmuir	[82]
	Cd^{2+}			48.02 mg/g		
Wood flour	Pb^{2+}	Tetraethylenepentamine	pH= 4.0; time= 3 hours; dosage= 1 g; temperature = 293 K; concentration= 300 mg/L	189.9 mg/g	Langmuir	[83]
amidoximated non-woven polyethylene-g-acrylonitrile fabric	Cu^{2+}	Acrylonitrile/ Hydroxyl amine hydrochloride	contact time = 72 hours; pH= 5.2; concentration= 500 ppm	74.62 mg/g	Langmuir	[84]
	Pb^{2+}		contact time = 72 hours; pH= 5.4; initial concentration= 500 ppm	107 mg/g		
	Cr^{6+}		contact time = 72 hours; pH= 1.5; initial concentration= 500 ppm	156.25 mg/g		
Silica	Mn^{2+}	Di (2-picolyl) amine	pH= 6.0; shaking time = 15 minutes; adsorbent dose= 20 mg	0.41 mg/g	Langmuir	[85]
Xanthan gum	Cu^{2+}	Ethylenediamine	contact time= 2 hours; pH= 5.0; dose= 3 g; initial concentration= 100 mg/L	46.95 mg/g	Langmuir	[86]
Micro-crystalline cellulose	Cd^{2+}	Sodium p-aminobenzoate	-	1.72 mmol/g	Langmuir	[87]
	Cu^{2+}			1.96 mmol/g		
	Ni^{2+}			1.88 mmol/g		

	Pb^{2+}			2.01 mmol/g		
	Zn^{2+}			1.93 mmol/g		
Polyacry-lonitrile Nanofiber Mats	Ag^{+}	Diethylenetriamine	time= 10 hours; concentration= 40 mg L^{-1}; pH= ≤ 7	12.23 mg/g	Freundlich	88
	Cu^{2+}		time= 5 hours; pH= ≤ 7; 40 mg/L (concentration)	30.40 mg/g	Langmuir	
	Pb^{2+}		pH= ≤ 7, time= 10 hours, 40 mg/L (concentration)	15.75 mg/g	Langmuir	
	Fe^{2+}		pH= ≤ 7, time= 5 hours, 40 mg/L (concentration)	5.42 mg/g	Langmuir	
Green seaweed	Cu^{2+}	Ethylenediamine	time= 30 minutes; pH= 5.6; dosage = 0.25 g; 170 mg/L (concentration)	5.27 mg/g	Langmuir	89
	Cd^{2+}		time= 30 minutes; pH= 6.3; dosage= 0.20 g; 170 mg/L (concentration)	2.12 mg/g	Langmuir	
	Pb^{2+}		time= 30 minutes; pH= 5.0; dosage = 0.20 g; = 90 mg/L (concentration)	2.16 mg/g	Freundlich	
Sugarcane bagasse	Cd^{2+}	Triethylenetetramine	pH= 5.5-6.0, equilibrium time= 40 minutes	313 mg/g	Freundlich	90
	Pb^{2+}		pH= 5.0-6.0, equilibrium time= 50 minutes	313 mg/g	Langmuir	
	Cu^{2+}		pH= 5.5-6.0, equilibrium time= 40 minutes	133 mg/g	Langmuir	
	Cd^{2+}	Ethylenediamine	pH= 6.5-7.5, equilibrium time= 40 minutes	164 mg/g	Langmuir	
	Pb^{2+}		pH= 5.0-6.0, equilibrium time= 50 minutes	189 mg/g	Langmuir	
	Cu^{2+}		pH= 5.5-6.0, equilibrium time= 40 minutes	139 mg/g	Langmuir	
Silica aerogels	Pb^{2+}	3-aminopropyl triethoxysilane	time= 48 hours, pH= 6.0, 200 mg/L (concentration); dosage= 1.6 g	45.45 mg/g	Langmuir	91
	Cd^{2+}		dose= 1.6 g, pH= 8.0, contact time= 48 hours, concentration= 100 mg/L	13.77 mg/g	Freundlich	

5. ADSORPTION KINETICS

Kinetic models provide information on the adsorption mechanisms and the potential rate-determining step[65, 92]. The models describe the adsorption system dynamics such as residence time, adsorption rate and mass transfer

parameters such as intra-particle diffusivity and external mass transfer[93]. This is achieved by using different kinetic models the one having the highest coefficient of correlation (R^2) values[94] is best-fit. Kinetic models such as intraparticle diffusion, pseudo second order kinetics, pseudo first order model, Elovich, Fractional power among others[95, 96, 97] are widely used.

5.1 Pseudo first order model

The model assumes diffusion as the rate limiting step which is physisorption in nature[98] and that occupation of metal ions is dependent on the number of unoccupied sites[99]. Its linearized equation[100] is expressed as:

$$\log (q_e - q_t) = \log q_e - {}^{k_{pf}}/_{2.303\ t}$$

where q_e (mg/g) is the adsorbed metal ions at equilibrium, q_t (mg/g) is the adsorbed metal ions at time (t) and k_{pf} is the rate constant. k_{pf} and q_e are calculated from slope and intercept of $\log (q_e - q_t)$ against time (t) plots.

5.2 Pseudo second order model

The model is based assumes chemisorption as the rate-determining step[5, 101]. The pseudo-second-order linearized equation is given as:

$$t/q_t = \frac{1}{k_2 q_e^2} + \left(\frac{1}{q_t}\right) t$$

where k_2 is the pseudo-second-order rate constant of adsorption. q_t and k_2 is obtained from a linear plot of t/q_t versus time (t) [102].

5.3 Weber–Morris intra-particle diffusion model

The model describes the diffusion of the metal ions within the pores of the adsorbent surfaces (intraparticle diffusion)[103]. The model is expressed by equation:

$$\log \%\, R = m \log t + \log K$$

where % R is the adsorbed metal ions adsorbed by percentage, t is the contact time, K and m are intra-particle diffusion constants[104]. The table below shows a summarized adsorption kinetic data on different amino-functionalized adsorbents:

Adsorbent	Metal ion	Modifying agent	Optimum parameters	Adsorption capacity		Rate constant (k)	Adsorption kinetic model	Reference
				Qe, exp (mg/g)	Qe, cal (mg/g)			
Fe_3O_4@ mesoporous SiO_2 core-shell	Pb^{2+}	3-methoxy salicylaldimine propyl triethoxysilane	pH= 5.0; adsorbent dose= 0.2 g	1.80	1.91	5.4×10^{-1} (g/mg/min)	Pseudo second order	[105]
		3-hydroxy salicylaldimine propyl triethoxysilane		1.64	2.22	2.0×10^{-2} (g/mg/min)	Pseudo second order	
Aspergillus niger Biomass	U^{6+}	Ethylenediamine	adsorbent dose= 0.2 g/L; Optimal pH= 5.0; contact time= 150 minutes; concentration=	-	2.46	1.22×10^{0} (g/mg/min)	Pseudo second order	[106]

			0.8 mg/L					
Fe_3O_4 nanoparticles	Cr^{6+}	1, 6-hexanediamine	Optimal pH= 3.0	24.25	28.25	2.0×10^{-3} (g/mg/min)	Pseudo second order	107
	Ni^{2+}		Optimal pH= 6.0	25.12	25.97	8.0×10^{-3} (g/mg/min)		
Attapulgite		3-aminopropyltriethoxysilane	contact time= 20 minutes; pH= 6.0	-	50.66	1.3×10^{-2} (g/mg/min)	Pseudo second order	108
	Cu^{2+}		contact time= 90 minutes; pH= 6.0	-	46.61	1.3×10^{-1} (g/mg/min)		
Kapok fiber	Pb^{2+}	Diethylenetriaminepentaacetic dianhydride	Optimum pH= 4.5; equilibration time= 2 minutes; concentration= 300 mg/L	298.0	302.1	2.90×10^{-2} (g/mg/min)	Pseudo second order	109
	Cd^{2+}		Optimum pH= 4.5; equilibration time= 2 minutes; concentration= 200 mg/L	153.4	153.8	2.24×10^{-1} (g/mg/min)	Pseudo second order	
	Cu^{2+}		Optimum pH= 4.5; equilibration time= 5 minutes; concentration= 200 mg/L	91.7	95.2	1.21×10^{-2} (g/mg/min)	Pseudo second order	
Lignin	Cu^{2+}	Diethylenetriamine	pH= 4.0	67.76	68.33	2.71×10^{-3} (g/mg/min)	Pseudo second order	110
	Pb^{2+}			49.61	51.93	1.09×10^{-1} (min^{-1})	Pseudo first order	
Polystyrene	As^{5+}	Diethylenetriamine	pH= 4.0; contact time= 2 hours	-	-	1.356×10^{-0} (g/ mmol/h)	Pseudo second order	111
Chitosan	Pb^{2+}	Polyaniline	concentration=40 mg/L; pH= 6.0; adsorbent dose= 0.15 g/L;	-	-	2.256×10^{-0} (mg/g/min)	Weber Morris	112
	Cd^{2+}			-	15.78	3.025×10^{-3} (g/mg/min)	Pseudo second order	

Maghemite nanoparticles	Pb^{2+}	Cysteamine	contact time= 100 minutes; pH= 5.0; adsorbent dose= 0.03 g	12.28	12.36	4.40×10^{-2} (g/mg/min)	Pseudo second order	[113]
	Hg^{2+}			24.96	24.67	4.0×10^{-3} (g/mg/min)	Pseudo second order	
	Cd^{2+}			11.92	12.04	8.0×10^{-3} (g/mg/min)	Pseudo second order	
	Ag^{+}			16.21	16.86	4.0×10^{-3} (g/mg/min)	Pseudo second order	
Zr-based metal-organic framework	Pb^{2+}	Ethylene diamine	pH= 5.6; agitation time= 200 minutes; adsorbent dosage= 2g/L; concentration= 300 mg/L	-	147.06	1.10×10^{-4} (g/mg/min)	Pseudo second order	[114]
	Cd^{2+}			-	123.46	3.10×10^{-4} (g/mg/min)	Pseudo second order	
	Cu^{2+}			-	120.48	2.40×10^{-4} (g/mg/min)	Pseudo second order	
Cassava starch	Cd^{2+}	Ethylenediamine	pH= 6.0; equilibrium time= 2 hours; concentration= 2.6 mmol/L	0.9160	0.7246	2.45×10^{-1} (g/mg/min)	Pseudo second order	[115]
Silica gel	Cu^{2+}	Nitrilotriacetic acid	pH= 6.0, time= < 2 minutes; concentration= 20 mg/L	20	20.01	2.46×10^{-1} (g/mg/min)	Pseudo second order	[116]
	Cd^{2+}		pH= 8.0, time= < 20 minutes; concentration= 20 mg/L	20	20.04	5.9×10^{-2} (g/mg/min)		
	Pb^{2+}		pH= 7.0, time= < 20 minutes; concentration= 20 mg/L	20	20.00	3.733×10^{-0} (g/mg/min)		

6. ADSORPTION THERMODYNAMICS

Optimization of temperature is parameter in describing adsorption of metal ions in a temperature controlled system[65, 117]. Thermodynamic parameters are used to describe the nature, feasibility and favorability of adsorption [118, 119]. The temperature data is analyzed by distribution constant (K_d), Van't Hoff equation and Gibb's free energy.

The distribution constant (K_d) can be expressed as[105]:

$$k_d = {}^{q_e}/_{c_e}$$

where q_e (mg/g) is the adsorbed metal ions at equilibrium and C_e (mg/L) is the residual metal ions adsorbed at equilibrium.

Gibb's free energy change (ΔG°) is calculated as follows[120]:

$$\Delta G° = -RT\ln k_d$$

where T is the absolute temperature (K) and R is the Molar gas constant (8.314 J $mol^{-1}K^{-1}$). The ΔG° values are used to describe the spontaneity of an adsorption [121]. Negative values of ΔG° shows that adsorption is spontaneous and positive values shows non-spontaneity nature of adsorption[122, 123].

The relationship between ΔG°, ΔH°, ΔS° and $\ln k_d$ is expressed by the equation[124]:

$$\ln k_d = \frac{\Delta S°}{R} - \frac{\Delta H°}{RT}$$

A plot of $\ln k_d$ verses ${}^{1}/_{T}$ yields a straight line with $-{}^{\Delta H^o}/_{R}$ (slope) and ${}^{\Delta S^o}/_{R}$ (intercept) which are used to calculate ΔH^o and ΔS^o respectively. ΔH° provides information on the endothermic or exothermic nature of adsorption [125]. Endothermic adsorption process is associated with increase in removal capacity as temperature is increased[126] vice versa is due to a decrease in removal capacity as the temperature is increased[127]. Positive ΔH° values can be used to describe adsorption nature (physisorption or chemisorption). That is, ≤ 40 kJ/mol (physisorption) and > 40 kJ/mol (chemisorption) [128]. The ΔS° provides information on randomness of surface sites during adsorbent-adsorbate interactions [129].

Adsorbent	Metal ion	Modifying agent	Optimum parameters	ΔG° (kJ/mol)		ΔH° (kJ/mol)	ΔS° kJ/mol/K	Reference
Attapulgite	Pb^{2+}	Ethylenediamine	pH= 4.0-6.0	300 K	-6.51	-45.30	-0.129	130
				308 K	-5.48			
				318 K	-4.18			
Chitosan	Pb^{2+}	4-aminobenzoic acid	pH= ≤ 7; contact time= 45 minutes; temperature= 45°C; dose= 6g/L; concentration= 0.5 mmol/L	298 K	-1.52	0.23	57.23	117
				303 K	-1.34			
				318 K	-4.45			
				328 K	-4.98			
	Zn^{2+}		pH= ≤ 7; contact time= 55 minutes; temperature= 45°C; dose= 6g/L; concentration= 0.5 mmol/L	298 K	-1.20	0.32	54.26	
				303 K	-2.32			
				318 K	-4.75			
				328 K	-6.43			
	Cu^{2+}		pH= ≤ 7; contact time= 45 minutes; temperature= 45°C; dose= 6g/L; concentration= 0.5 mmol/L	298 K	-1.25	0.26	64.78	
				303 K	-1.65			
				318 K	-3.97			
				328 K	-4.91			
	Ni^{2+}		pH= ≤ 7; contact time= 45 minutes; temperature= 45°C; dose= 6g/L; concentration= 0.5 mmol/L	298 K	-2.31	0.20	67.12	
				303 K	-6.92			
				318 K	-8.78			
				328 K	-9.56			
	Cd^{2+}		pH= ≤ 7; contact time= 55 minutes; temperature= 45°C; dose= 6g/L; concentration= 0.5 mmol/L	298 K	-2.58	0.27	68.45	
				303 K	-4.41			
				318 K	-8.54			
				328 K	-9.43			
Bacterial cellulose	Cu^{2+}	Sulphamate	Initial concentration= 600	293 K	1.16	43.16	0.15	
				298 K	0.43			
				308 K	-0.75			
	Pb^{2+}			293 K	-0.002	57.65	0.20	

			mg/L	298 K	-0.99			[131]
				308 K	-1.75			
	Cd^{2+}			293 K	-0.25			
				298 K	-1.47	71.76	0.25	
				308 K	-2.60			
Bentonite	Cu^{2+}	Tetraethylenepentamine	-	298.15 K	-17.9			[132]
				308.15 K	-19.9			
				318.15 K	-22.4	47.1	0.22	
				328.15 K	-24.3			
	Ni^{2+}			298.15 K	-17.5			
				308.15 K	-19.8			
				318.15 K	-21.4	42.7	0.20	
				328.15 K	-23.6			
	Cd^{2+}			298.15 K	-14.3			
				308.15 K	-16.0			
				318.15 K	-17.6	35.6	0.17	
				328.15 K	-19.4			
Graphene oxide	Cd^{2+}	3-aminopyrazole	Optimal pH= 8.8; dose= 10 mg; concentration= 50 ppm	298 K	-11.91			[133]
				308 K	-16.89	13.27	0.48	
				318 K	-21.61			
	Hg^{2+}		Optimal pH= 8.3; dose= 10 mg; concentration= 50 ppm	298 K	-6.05			
				308 K	-6.89	19.06	0.08	
				318 K	-7.73			
	As^{3+}		Optimal pH= 7.6; dose= 10 mg; concentration= 50 ppm	298 K	-1.51			
				308 K	-1.96	11.94	0.05	
				318 K	-2.44			
Cellulose (Commercial)	Hg^{2+}	N,N -dimethyl benzal aniline	pH= 5; dosage= 0.3 g/L	308 K	-148.52			[134]
				313 K	-184.76			
				318 K	-219.43	-15.38	0.54	
				323 K	-255.11			
Kaolinite clay	Pb^{2+}	Ethylenediamine	Optimal pH= 5.5; dose= 0.1 g	298 K	3.23			[122]
				313 K	0.35	-0.45	0.008	
				323 K	0.31			
				333 K	0.29			
	Cd^{2+}		Optimal pH= 4.5; dosage= 0.1 g	298 K	1.66			
				313 K	1.17	-0.14	-0.00	
				323 K	1.02			
				333 K	0.70			
Chitosan	Cu^{2+}	Ethylenediaminetetraacetic acid	pH= 5.5; contact time= 40 minutes; concentration= 200 mg/L	303 K	-12.33			[135]
				313 K	-12.82	2.73	0.05	
				323 K	-13.32			
D301 resin	Cu^{2+}	Iminodiacetic acid	pH=5.0	293 K	-9.35			[121]
				298 K	-9.63			
				303 K	-9.95	8.11	0.059	
				308 K	-10.22			
				313 K	-10.54			
	Pb^{2+}			293 K	-9.46			
				298 K	-9.75			
				303 K	-10.02	6.68	0.055	
				308 K	-10.30			
				313 K	-10.57			
	Cd^{2+}			293 K	-9.42			
				298 K	-9.69			
				303 K	-10.01	8.12	0.060	
				308 K	-10.28			
				313 K	-10.62			
Biomass ash	Cd^{2+}	3-	Optimum pH= 5.0; contact					

(wheat stem, groundnut shell, maize straw, cotton stalk)		aminopropyltriethoxysilane	time= 90 minutes; concentration= 50 mg/L	30 ^{o}C	-2.31	39.35	0.14	136
				45 ^{o}C	-3.35			
				60 ^{o}C	-5.63			
Chicken feathers	Cu^{2+}	Ethylenediamine	pH= 6.0; dosage = 9.0 g/L; contact time= 60 minutes; concentration= 20 mg/L	303 K	-8.84	4.57	0.044	137
				313 K	-9.26			
				323 K	-9.72			
	Co^{2+}		pH= 6.0; dose= 4.0 g/L; contact time= 12 minutes; concentration= 20 mg/L	303 K	-10.27	4.57	0.049	
				313 K	-10.79			
				323 K	-11.25			
	Ni^{2+}		pH= 6.0; adsorbent dose= 7.0 g/L; contact time= 24 minutes; concentration= 20 mg/L	303 K	-11.56	100.93	0.101	
				313 K	-12.28			
				323 K	-13.56			
	Fe^{2+}		pH= 6.0; dose= 9.0 g/L; contact time= 40 minutes; concentration= 20 mg/L	303 K	-8.41	41.48	0.041	
				313 K	-8.82			
				323 K	-9.23			
Corncob	Cd^{2+}	Polyacrylamide	pH= 7.0; dosage= 5 g/L; temperature= 30 ^{o}C	283 K	-10.64	0.72	0.04	138
				293 K	-11.03			
				303 K	-11.47			
				313 K	-11.86			
				323 K	-12.24			

7. CONCLUSION

Its clear from the reports reviewed that amino-functionalized adsorbents are emerging as excellent candidates for decontamination of contaminated water. The parameters of adsorbent dose, temperature, pH, time and concentration have been reported to greatly influence the removal of heavy metal ions. However, their use require further investigation in the direction of modelling validation, adsorbent regeneration and metal ion recovery.

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