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Overview of Adsorption Isotherm studies of heavy metals from wastewater using coal fly ash

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Abstract: Heavy metals pollution in wastewater is a great environmental challenge. Several techniques and materials have been recently proposed in order to solution this problem, such as the adsorption process. Adsorption technology is currently being used extensively for the removal of heavy metals from aqueous solutions because it is a cleaner, more efficient and cheaper technology. Three kinetic mechanisms are used for adsorption on heterogeneous substrates as is the fly ash: pseudo-first order, pseudo-second order and intra-particle diffusion. The thermodynamic parameters such as the free energy $\Delta G°$, enthalpy $\Delta H°$ and entropies $\Delta S°$ of showed that the adsorption process is suitable, spontaneous and endothermic.

Key words: Heavy metals, fly ash, adsorption, kinetics, thermodynamics

INTRODUCTION

Fly ash (FA) is one of the major industrial wastes generated from power stations that cannot be cheaply disposed off. Recent research efforts have consequently focused on developing ways to make use of FA in applications that are friendly to the environmentally benign. Apart from its limited applications in cement and concrete industries, FA alternative use/reuse in environmental study takes advantage of its reasonable adsorptive property for water treatment. Limited studies have highlighted the need to improve FA adsorption capacity[1].

Conventional methods including, reverse osmosis[2], electrodialysis[3], ion exchange[4], chemical precipitation[5], ultra filtration[6] and adsorption are used for removal of heavy metal ions from wastewater. The adsorption method, among all above mentioned processes[7]. It is the most preferable one because it is economically advantageous, highly efficient and applicable[8]. Adsorption process gives an attractive alternative for the treatment of contaminated water. It remains an innovative and effective alternative treatment for heavy metals removal from wastewater[9]. Adsorption is efficient and reliable method for removal of heavy metals. It also

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gives high adsorption capacity and inexpensive method[10]. Fly ash is a by-product of coal combustion occurring in thermal power plant processes. The fact that coal is used as a fuel in most industries causes FA to be produced in higher amounts. It has proven it's and suitability as adsorbents to eliminate heavy metals from wastewater[11].

Adsorption Isotherm - The descriptions of adsorption behaviors are usually provided by mathematical models known as the adsorption isotherm models[12]. An adsorption isotherm equation is an expression of the relation between the amount of solute adsorbed and the concentration of the solute in the fluid phase[13]. Adsorption equilibrium is important and effective ways to research the adsorption process[14]. The Langmuir adsorption model is based on the assumption that sorption occurs at specific homogeneous sorption sites within the adsorbent and intermolecular forces decrease rapidly with the distance from the sorption surface. The model is also based on the assumption that all the sorption sites are energetically identical and sorption occurs on a structurally homogeneous sorbent[15]. The Freundlich adsorption isotherm is commonly used to describe the adsorption characteristics of a heterogeneous surface[16]. It is a nonlinear sorption model proposing a monolayer sorption with heterogeneous distribution of active sites involving interaction between adsorbed molecules[17].

Adsorption kinetics

The kinetic investigations are important for adsorption studies since they can predict the potential rate controlling step and the mechanism of adsorption reactions. For the investigation of adsorption mechanisms, in recent years, adsorption mechanisms involving kinetics-based models have been reported. Numerous kinetic models have described the reaction order of adsorption systems based on the solution concentration[18]. Kinetics of adsorption data is analyzed using different kinetic models, such as pseudo-first order, pseudo-second order and intraparticle diffusion. Various kinetic models including Morris- Weber, Lagergren and Elovich model have been utilized due to their consistency with the experimental adsorption data. It is obtained for adsorption of heavy metals onto fly ash. No mass transfer (either external or internal) resistance was expected to happen to the overall adsorption process, Therefore kinetic can be studied based upon the concentration of residual metal ion in the solution. Studying kinetic of adsorption helps us to find out the solute uptake rates. In which clearly, can be used to control the residence time of adsorbate uptake at solid-solution interface including the diffusion process[19].

Pseudo-first-order kinetic

In 1898, Lagergren presented a first-order rate equation to describe the kinetics of liquid-solid phase[20]. This phase declares that the rate of change that occurred in adsorbate uptake with the passage of time is directly proportional to the difference in the saturation concentration and the rate of solid uptake with time. The Lagergren equation is the most commonly used rate equation in liquid phase sorption. The pseudo-first-order equation is described in Equation (1) and is often used for estimating k_1 considered to be a mass transfer coefficient in design calculations[21].

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$$\log(q_e - q_t) = \frac{log q_e}{2.303} - k_1 t \qquad (1)$$

Where q_e and q_t are the amounts of metal ions adsorbed (mg/g) at equilibrium and at time t (min), respectively, and k_1 (min^{-1}) is the rate constant of first-order adsorption. The values of k_1 are calculated using the plots of log ($q_e - q_t$) versus t[22]. Table-1 showed the pseudo- first order

Table - 1: Pseudo first order

Metal	k_1 (min^{-1})	q_e (mg g^{-1})	R^2	References
Cu	0.087	2.8	0.79	15
Pb	0.034	0.834	0.960	19
Ni	0.007	0.0476	0.797	20
Cu	0.0378	70.5	0.9588	21
Mn	0.0496	41.4	0.9475	,,
Ni	0.0361	54.3	0.9702	,,
Pb	0.0198	108.9	0.3079	,,
Zn	0.0319	66.0	0.7874	,,
Pb	0.02	6.3×10^{-7}	0.652	22
Cd	0.00759	0.10897	0.9989	23
Zn	0.000207	0.731	0.9857	,,
Cr	0.0055	1.5966	0.2027	24
Cr	0.7823	8.39	0.8847	25
Hg	-	-	0.9629	26
Pb	-	-	0.9555	,,
Zn	0.055	35.73	0.6427	27
Cu	-	-	0.61	28
Zn	-	-	0.45	,,
Hg	0.03	0.28	0.94	29
As	0.005	11.5	0.8306	30
Cu	1.3	36.36	0.893	31
Ni	0.38	28.18	0.961	,,

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Cu	0.30	2.07	0.9756	32
Fe	0.16	3.30	0.9374	,,
Mn	0.12	4.96	0.9587	,,
Zn	0.14	4.23	0.9689	,,
Cr	0.199	3.56	0.994	33

Pseudo Second order kinetic

The Pseudo second order kinetic equation, developed by Ho and McKay[34]. It is based on the adsorption capacity of the solid phase. If the rate of sorption is a second-order mechanism, the pseudo second-order equation indicates chemisorptions, which implies a strong electrostatic interaction between the negatively charged surface and metal ions.

$$\frac{dq_t}{dt} = k^2 (qe - qt)^2 \qquad (2)$$

Where q_e and q_t are the adsorption capacity (mg g-1) of metal ions at equilibrium and at a time t, k_2 is the equilibrium rate constant for the second-order adsorption (g/mg^{-1} min^{-1}). This model is based on the assumption that the rate limiting step may by a chemical adsorption involving the valence forces through sharing or exchange of electron between the adsorbent and the adsorbate. The activation energy of adsorption is evaluated with the pseudo-second-order rate constants. R^2 values are obtained from the linear plot of t/qt versus 1/qe^2. Table (2) showed the pseudo - second order

Table - 2 : Pseudo second order

Metal	K_2 (g mg^{-1}min$^{-1)}$	q_e (mg g^{-1})	R^2	References
Cd	0.000869	16.583	0.9964	11
Cu	0.104	8.31	0.99	15
Pb	0.065	2.04	0.999	19
Ni	2.0216	0.0714	0.998	20
Cu	1538.89	70.5	0.9907	21
Mn	137.77	41.4	0.9996	,,
Ni	631.27	54.2	0.9980	,,

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Pb	141.78	108.9	0.9999	,,
Zn	27.80	66.0	0.9996	,,
Pb	1.97×10^{3}	2.25×10^{-5}	0.999	22
Cd	0.2195	0.8407	0.9999	23
Zn	0.3578	0.8813	0.9999	,,
Cr	-2.3359	4.027	0.999	24
Cr	0.7726	9.514	0.9999	25
Hg	-	-	0.9906	26
Pb	-	-	0.871	,,
Zn	0.0395	14.28	0.9661	27
Cu	15.0	-	0.999	28
Zn	6.4	-	0.997	,,
Hg	54.0	0.73	1	29
As	0.0008	24.06	0.9979	30
Cu	12.33	2.07	1.000	32
Fe	0.58	3.29	0.9918	,,
Mn	0.28	4.98	0.9978	,,
Zn	0.38	4.24	0.9999	,,
Cr	0.156	8.22	0.999	33
Pb	10.8	-	1.000	34
Cd	3.6	-	0.999	,,
Cu	0.0187	15.8983	0.9997	35
Cu	0.0244	20.0401	0.9998	,,
Cu	0.0513	20.3666	1.0	,,
Cd	0.0082	29.58	0.9371	36
Ni	4.12232	22.03	1.0000	,,

Intra-particle diffusion kinetic

The intra-particle diffusion is kinetic model proposed by Weber and Morris. The adsorption process

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requires a multi-step approach involving the transport of solute molecules from the aqueous phase to the surface of the solid particles, followed by diffusion of the solute molecules into the interior of the pores, which is likely to be a slow process.

$$q_t = k_{id} (t)^{1/2} + C_i \qquad (3)$$

Where k_{id} is the rate parameter of i stage (mg/g $h^{1/2}$), calculated from the slope of the straight line of qt versus $t^{1/2}$. C_i is the intercept of i stage, indicating the thickness of boundary layer, that is, the larger the intercept, the greater the boundary layer effect is. For intra-particle diffusion, q_t versus $t^{1/2}$ will be linear and if the plot passes through the origin, then the rate limiting process will only be due to the intra-particle diffusion[37]. Table-3 showed the Intra - Particle Diffusion

Table - 3 : Intra - Particle Diffusion

Metal	k_{id} (mg $g^{-1}min^{-1/2}$)	A	R^2	References
Cd	0.0046	0.8413	0.8365	2
Pb	0.10	-	0.789	19
Ni	0.003	-	0.849	20
Cu	4.7038	16.6355	0.8662	21
Mn	0.9998	30.060	0.8075	,,
Ni	2.9666	20.952	0.7178	,,
Pb	1.5039	91.507	0.4227	,,
Zn	2.3507	39.680	0.5535	,,
Cd	6.3841	0.0476	0.9996	23
Zn	7.5927	0.0253	0.9369	,,
Hg	-	-	0.9810	26
Pb	-	-	0.9739	,,
Zn	0.7179	6.4466	0.8979	27
Cu	0.60	3.7	0.999	28
Zn	1.61	2.2	0.998	,,
As	0.678	5.40	0.6637	30
Pb	0.6	4.2	1.000	34
Cd	1.6	2.7	0.997	,,
Cd	0.952	10.4	0.9504	38

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Cd	5.069	63.414	0.844	,,
Cu	5.964	22.128	0.928	,,

Elovich kinetic

The Elovich equation, which applies to heterogeneous surfaces (Chien and Clayton, 1980), was also applied in an effort to better describe the chemisorptions process. It is generally expressed as:

$$q_t = 1/\beta \text{ In } \alpha\ \beta + 1/\beta \text{ In } t \qquad (4)$$

Where: α is the initial adsorption rate (mg g^{-1} h^{-1}) and β is related to the extent of surface coverage and the activation energy for chemisorption (g^{-1} mg).

Table - 4 : Elovich kinetic

Metal	α_2(mg/g)	β(g/mg)	R^2	References
Cd	0.623	0.3037	0.9571	11
Cr	9.041	0.1263	0.9998	25
Zn	0.781	2.32×10^5	0.7078	27
Cu	375	0.31	0.907	28
Zn	160	0.18	0.915	,,
Hg	60.7	16.45	0.94	29

Thermodynamic study

Thermodynamic study tells the feasibility, nature and spontaneity of the reaction. All parameters calculated by thermodynamic equation decide the process behavior. The thermodynamic parameters, which characterize the equilibrium of a system, are the Gibbs free energy (ΔG°), the enthalpy (ΔH°) and the entropy (ΔS°). The apparent distribution coefficient (K_d) of the adsorption is defined as:

$$\Delta G^\circ = -RT \ln K_0 \qquad (5)$$

Where K_0 is equilibrium constant. Gibbs free energy change is directly related to change in enthalpy and change in entropy.

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Temperature is an important parameter in a thermodynamic study. The experiments showed that the adsorption capacity decreases with decrease in the solution temperature. The decrease in the rate of adsorption with the temperature drop may be attributed to the weakening of the adsorptive forces between the active sites of the adsorbents, and adsorbate species and the adjacent molecules of the adsorbed phases. The variation in the extent of adsorption with respect to temperature has been explained on the basis of thermodynamic parameter, like change in enthalpy ΔH^0,change in entropy ΔS^0 and change in Gibb's free energy ΔG^o, were determined using the following equation (6),(7) and (8):

$$\Delta G^o = -RT \ln K_1 \quad (6)$$

$$\Delta H^0 = E_a - RT \quad (7)$$

$$\Delta S^0 = \frac{\Delta H^0 - \Delta G^o}{T} \quad (8)$$

The activation energy E_a can be calculated using the slope and intercept of the Arrhenous plot of $\ln k_L$ versus I /T.

Table-5: Thermodynamic parameters for the adsorption of metals

Metal	ΔH°(Kcal/mol)	ΔS°(Kcal/mol)	ΔG°(Kcal/mol)	Temp. (K)	References
Cd	2.43	13.045	-1.53	30°	23
			-1.64	40°	
			-1.79	50°	
Zn	2.85	15.805	-1.93	30°	
			-2.09	40°	
			-2.25	50°	
Zn	7.04	47.7	-6.47	0 C	39
	7.04	47.7	-6.85	15	
	7.04	47.7	-7.1	23	
	7.04	47.7	-7.41	40	
	7.04	47.7	-7.76	55	
Cd	-15.00	0.024	-22.24	303K	40
	-15.00	0.010	-18.02	313K	
Cu	67.64	237.5	-1.95	293	41
			-6.4	308	
			-9.11	318	

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Mn	7.34	28.28	0.72	298	42
	4.28	13.49	-11.35	333	
	-	-	0.43	303	
			0.31	308	
Zn	61.46	213.93	-1.22	293	41
As	30.6	0.123	-6.04	298	30
			-7.01	308	
			-8.67	318	
			-9.59	328	
Cu	53834.1	211.4	-9118.7	298	15
			-13468.4	318	
			-16493.0	333	
Cu	18200.6	67.1	-1855.9	298	15
			-3001.1	318	
			-4241.0	333	
Cu	200.7498	51.6557	-6.4551	20°c	35
			-10.3853	30°c	
			-12.7672	50°c	
Zn	26.4	101	-3.62	25°c	43
	24.9	95.9	-4.12	30°c	
	-	-	-5.08	40°c	
Cd	45.5	170	-5.29	25°c	43
	29.7	118	-6.14	30°c	
	-	-	-7.32	40°c	
Pb	40697.03	146.8252	-1385.76	35°c	44
			-2278.00	45°c	
			-2692.38	55°c	

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Conclusion

As low cost alternatives, raw and activated fly ashes are considered inexpensive and environmental-friendly material. Adsorption is a reliable and inexpensive method. It gives high adsorption capacity. This research highlights that fly ash material, a hazardous industrial waste, has a great potential in water treatment application. Kinetic and thermodynamic parameters were determined to the adsorption mechanism of each heavy metals on to fly ash samples. Three kinetic mechanisms are used for adsorption on heterogeneous substrates as is the fly ash: pseudo-first order, pseudo-second order and intra-particle diffusion. Thermodynamic parameters (ΔG°, ΔH° and ΔS°) were also evaluated for the adsorption showing that the adsorption process is suitable, spontaneous and endothermic.

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