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Adsorption Kinetics of Pb(II) and Zn(II) ions unto Carbonized Groundnut Shells and Maize Cobs

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Abstract

Maize cobs and groundnut shell adsorbents from agricultural by-products were carbonized and investigated for adsorption of two selected metals, Lead (Pb II) and Zinc (Zn II) from aqueous solutions of these metals. Kinetic studies of the adsorption processes were investigated employing Elovich and the intraparticle diffussion models. The results showed that, the adsorption capacities of these comparatively cheaper adsorbents and their efficiency for the removal of Pb(II) and Zn(II) ions from aqueous solution present them suitable for use in place of the costly commercial activated carbons.

Keywords: Heavy metal ions, Maize cob, Groundnut shell, Adsorption kinetics

Introduction

Human activities create a vast amount of various wastes and pollutants, the release of these materials into the environment sometime cause serious health problems. Waste is considered as any substance, solution, mixture or article for which no direct use is envisaged but which is transferred for processing, dumping, elimination by incineration or other methods of disposal. Amongst all the classes of solid waste that pose the greatest threat to life, due to the potential of polluting the terrestrial, aquatic and aerial environments, heavy metals have been of great concern in the last decades because of their health hazards to man and other organisms when accumulated within biological systems[1]. The presence of these metals in the environment is of major concern because of their toxicity, non-biodegradable nature, bio-accumulating tendency, threat to human life and the environment [2, 3, 4]. The rapid growth in global population and urbanization has driven an exponential increase in industrial activities and haphazard rapid urbanization, which is accompanied by an increase in the amount of industrial wastes being discharged into the environment [3,5]. Hence the increase in heavy metals such as Cadmium, Mercury, Lead, Copper, Zinc, Nickel, Chromium, etc., posing significant risk to soil, water and human health [1,3,4]. They constitute pollution problems to the environment and they are toxic to man and aquatic life [6]. One of the most important problems is their incorporation into the food chain. As a result their accumulation here can be more than that in water and air. The contaminated food can cause poisoning in humans and animals. Although some heavy metals are necessary for the growth of plants, over the threshold, they become poisonous for both plants and microorganisms [7].

The heavy metal ions are stable and persistent environmental contaminants since they cannot be degraded and destroyed. They are also harmful to aquatic life and water contaminated by toxic metal ions remains a serious problem for human health [8]. Another important risk concerning heavy metal contamination is the accumulation of these metals in the soil as a result of adsorption, chemical reaction and ion exchange of soil [7]. Removal of heavy metals from their sources like wastewater and industrial wastes has become a very important environmental issue. The process of adsorption is considered one of the suitable for removal of contaminants from water [9]. Numerous treatment methods have been employed for the removal of heavy metals from industrial effluents. Conventional methods usually employed for the cleanup of these wastes are chemical precipitation, sedimentation, filtration, flocculation, neutralization, ion exchange, electrodialysis, reverse osmosis, oxidation, solvent extraction and adsorption [3, 6, 10, 11, and 12]. Adsorption is a surface phenomenon that occurs when a gas or liquid solute accumulates on the surface of a solid or liquid forming a molecular or atomic film. Adsorption has been described as an effective separation process for treating industrial and domestic effluents. It is widely used as effective physical method of separation in order to eliminate or lower the concentration of a wide range of dissolved pollutants (organics or inorganics) in the effluent [13]. The process of adsorption implies the presence of an “adsorbent” solid that binds molecules by physical attractive forces, ion exchange and chemical binding. It is advisable that the adsorbent is available in large quantities, easily regenerable and cheap [14]. Adsorption of these heavy metals on conventional adsorbents such as activated carbon have frequently been employed, but just like the other conventional adsorbents, the methods are often expensive and difficult to maintain due to high capital and operational costs, as well as extra cost of treating the resultant sludge generated before disposal [3,6]. A search for low-cost and easily available materials with high adsorptive capacities has led to the investigation of materials of biological origin as potential metal biosorbents [3,10]

The use of biomaterials as biosorbents for the treatment of waste water as a potential alternative to the conventional treatment method has been confirmed in separate studies [15]. The study of biosorption is of great importance from an environmental point of view, as it can be considered as an alternative technique for removing toxic pollutants from wastewater [10]. Various biomass materials and agricultural by-products have been utilized in the removal of toxic heavy metals from wastewater such as, maize cobs, cassava waste, rice husks, maize husks, sunflower stalk, coconut fibre, sugarcane bagasse, sawdust, sago waste, peanut skin, *Medica sativa* (Alfalfa), reed plant, banana peel, tree bark, apple waste, teawaste, etc.[2,3,5,8,16,17,18,19,20,21]. Metal biosorption is a rather complex process affected by several factors. Mechanisms involved in the biosorption include chemisorption, complexation, adsorption-complexation on surface and pore, ion exchange, microprecipitation, heavy metal hydroxide condensation onto the biosurface and surface adsorption [10]. The choice of an organic adsorbent usually depends on a number of factors which include availability, location, locality, seasonality, cost-effectiveness, etc. Groundnut shells and maize cobs are renewable/ biodegradable agricultural wastes available abundantly in large quantity at little or no cost. Their disposal form litters and constitute environmental pollution mostly in North-central and North-eastern parts of Nigeria, other regions and nations that cultivate maize and groundnut. This constitutes a major obstacle to sustainable environment.

The piece of research aims to assess the effectiveness and kinetics of commonly available agricultural by-products in the removal of toxic metals from their sources before discharging into the environment to safeguard the biotic factors of the eco-system. Two heavy metals ions Zn(II) and Pb(II) were selected and used for the study of adsorption kinetics of groundnut shells and maize cobs. Zn is an essential element to man, being a cofactor for many enzyme systems. It has been reported to competitively inhibit Pb uptake in cells. Toxic levels of Pb in man have been associated with encephalopathy, seizures and mental retardation [22].

Materials and Methods

Sample Collection and Preparation

The Groundnut shells were collected from a local farm in Janruwa, Kaduna State, North-east Nigeria. They were air-dried, crushed in a mortar and sieved through 600µm mesh. The Maize cobs were collected from a local farm in Lafia, Nasarawa State, North-central Nigeria. They were cut into small pieces, air-dried, crushed in a mortar and sieved through 600µm mesh to get fine particles of high surface area, for effective adsorption.

Carbonization of the Adsorbents

The groundnut shells and maize cobs were carbonized as described by Ademiluyi, P.T, *et al* [23] at a temperature of 300 ^{0}C. The carbonization of un-weighed groundnut shells and maize cobs were carried out in a muffle furnace. The samples were distributed into crucibles and heated in a muffle furnace to a temperature of 300 ^{0}C for 1 h after which the charred products were allowed to cool to room temperature and kept in a dessiccator for use.

Characterization of the Adsorbents

Physical characteristics namely ash content (%), moisture content (%), pH, of the adsorbents used in this study were determined.

Chemical Activation

The carbonized groundnut shells and maize cobs were steeped in 0.1M K_2CO_3 solution in 250cm^3 beaker for about 2-3hrs, the chemically activated samples were filtered and washed with distilled water. The pH of the activated sample was adjusted to fall between 6-7 using 0.1M HCl solution. The samples were then dried in an oven at 100°C for 3 h. The final products were kept in a tight container, ready for use.

Preparation of Metal Solutions

All metal salts and chemical reagents were of analytical grade (Aldrich U.K). Standard stock solutions of Zn^{2+} and Pb^{2+} were prepared to the required concentrations in mg/L by dissolving the appropriate amount of $Pb(NO_3)_2$ and $ZnSO_4$ in de-ionized water.

Batch Adsorption Studies

The batch experimental procedure was carried out as described by Donbebe *et al* [24]. The batch experimental procedure was applied to determine the contact time required for equilibrium sorption of Pb^{2+} and Zn^{2+} ions by putting 0.1g of chemically activated carbon of maize cobs into several flasks. 0.003g/L solution of Pb^{2+} (from $Pb(NO_3)$ and 0.0062g/L of Zn^{2+} (from $ZnSO_4$) were added respectively. Agitation was carried out with the aid of an electric shaker at a constant mixing speed for time interval of 5,10,20,40 and 60 min. At the end of each time interval, the mixture was removed and filtered rapidly. The metal ion concentrations were determined using the Atomic Adsorption Spectrometry (AAS). The amount of the metal adsorbed by the maize cobs is the difference between the initial and the final concentration of the solutions.

Results and Discussion

Adsorbent Characterization

Table 1: Physico-chemical properties of Groundnut shell and Maize cob

Parameter	Groundnut shell	Maize cob
Moisture content (%)	7.52	10.38
Total ash (%)	3.71	8.66
pH	6.4	7.3

Table 2: Percentage of Pb (II) ion adsorbed at concentration of 148.9768 ppm

	Time (min)	Amount adsorbed	% adsorbed
Maize cob	5	69.0460	46.35
	10	72.6940	48.79
	20	100.7069	67.59
	40	114.0386	76.55
	60	123.6332	82.98
Groundnut shell	5	73.2902	49.19
	10	78.7889	52.89
	20	108.5230	72.85
	40	126.1448	84.69
	60	134.2438	90.11

Table 3: Percentage of Zn (II) ion adsorbed at concentration of 5.8083ppm

	Time (min)	Amount adsorbed	% adsorbed
Maize cob	5	2.4998	43.04
	10	3.5227	60.65
	20	4.0766	70.19
	40	4.4780	77.09
	60	4.6920	80.78
Groundnut shell	5	2.7711	47.71
	10	3.9575	68.14
	20	4.3423	74.76

Table 3: Percentage of Zn (II) ion adsorbed at concentration of 5.8083ppm - continued

	40	4.6865	80.69
	60	4.7970	82.59

Figure 1: Effect of contact time for the adsorption of lead (II) ion

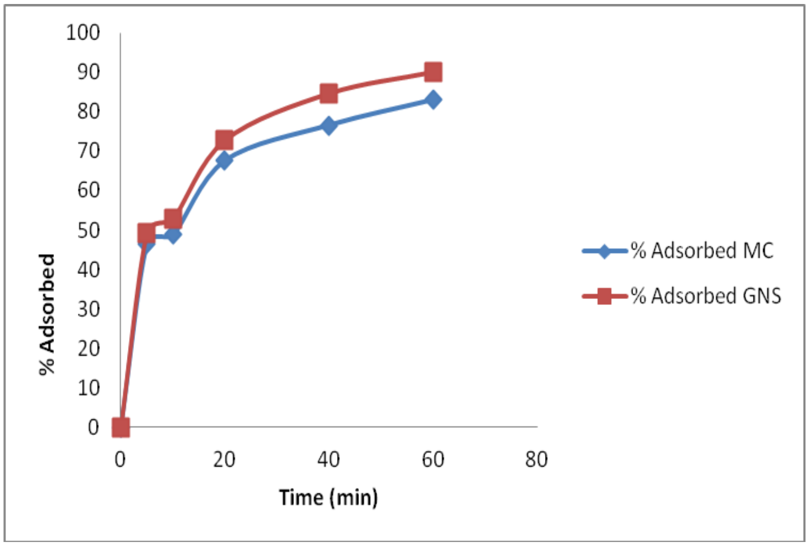
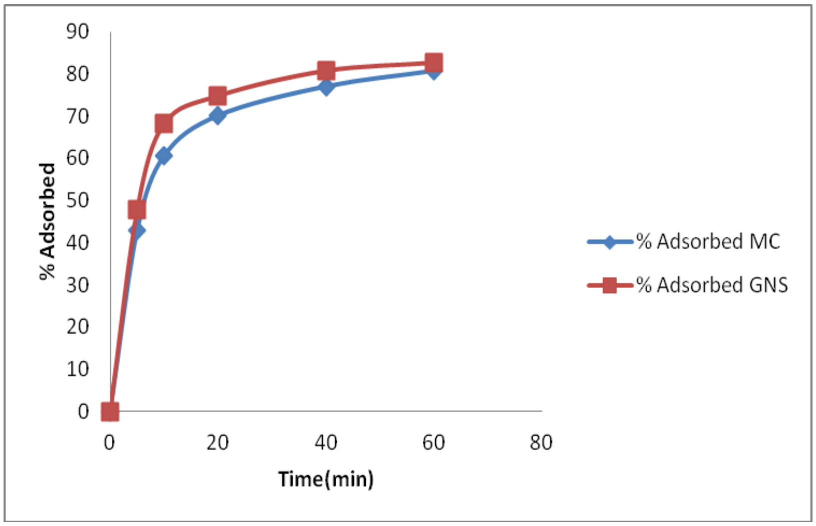


Figure 2: Effect of contact time for the adsorption of zinc (II) ion



The physico-chemical properties of groundnut shell and maize cob such as moisture content, total ash and pH were determined and shown on table 1. The moisture content of substance tells about the biodegradability of that substance. From the characterization results presented, the maize cob has a better biodegradability attribute, however, it compares less to groundnut shell in adsorption, likewise the total ash content which is representative of the inorganic content of a substance.

From table 2, the percentage of Pb^{2+} ion adsorbed is a function of contact time. The % adsorbed by both adsorbents increases with an increase with contact time. This also agrees with their adsorption capacity q_e values. However, the groundnut shell shows higher adsorption capacity with 90.11% at the same concentration due to its low moisture content.

Table 3 also shows a similar result. The percentage of Zn^{2+} ion adsorbed increases with increase in contact time, hence the groundnut shell shows high adsorption capacity with 82%.

Similar trend has been reported by Abechi *et al* [19] in which the adsorption capacity increases as the initial concentration of the metal ion increases. Generally, the percentage of Pb^{2+} ion adsorbed ranges from 49.35 to 90.11% and that of Zn^{2+} ion ranges from 43.04 to 82.59%. This is a pointer to the effectiveness of the prepared activated carbon for use on industrial scale.

The result of contact time experiment is presented in figure 1 and figure 2 above. There was an increase in the amount of metals adsorbed which increased gradually as the contact time increases. The curves show that groundnut shell has the highest adsorption capacity for both metals ions.

Kinetics of Adsorption

The study of adsorption kinetics is significant as it provides valuable insights into the reaction path ways mechanism of the reactions. Any adsorption process is normally controlled by three steps:

1. Transport of the solute from bulk solution to the film surrounding the adsorbent.
2. From the film to the adsorbent surface.
3. from the surface to the internal sites followed by binding of the adsorbent species into the active sites [25]**.**

The study of adsorption dynamics describes the solute uptake rate and evidently this rate controls the contact time of the adsorbate uptake at the solid-solution interface [6]. The kinetics of Zn^{2+} and Pb^{2+} adsorption on maize cob and groundnut shell were analyzed using Elovich [26, 27] and intraparticle diffusion [28] kinetic models. The conformity between experimental data and the model predicted values was expressed by the correlation coefficients (R^2 values close or equal to 1). A relatively high R^2 value indicates that the model successfully describes the kinetics of the adsorption [6].

Elovich Equation

The applicability of the Elovich equation to the sorption process was also tested. The Elovich equation is generally expressed as Chien *et al* (26).

$$dq_t/ dt = \alpha \exp (-\beta q_t) \quad (1)$$

Where, α is the initial sorption rate (mg.g-1. min-1); β is desorption rate constant (mg.g-1 min-1) during any one experiment. To simplify the Elovich equation, Chien *et al*, assumed αβt >>1 and by applying the boundary conditions $q_t = 0$ at $t = 0$ and $qt = q_t$ at $t = t$, Equation (2) becomes [28]

$$q_t = (1/\beta) \ln (\alpha\beta) + (1/\beta) \ln (t) \quad (2)$$

Thus, a plot of q_t vs. ln (t) should yield a linear relationship with a slope of (1/β) and an intercept of (1/β) In(αβ), if the sorption process fits the Elovich equation.

Intraparticle Diffusion Rate Equation

The sorption rate is shown to be controlled by several factors including the following processes;

(i) Diffusion of the solute from the solution to the film surrounding the particle.
(ii) Diffusion from the film to the particle surface (external diffusion).
(iii) Diffusion from the surface to the internal sites (surface or pore diffusion)
(iv) Uptake which can involve several mechanisms, such as physico chemical sorption, ion exchange, precipitation or complexation [30,31]. Due to rapid stirring during the batch experiment there is a possibility of transport of heavy metal ions from the bulk into pores of the adsorbent as well as adsorption at the outer surface of the adsorbent. The rate-limiting step may be either film diffusion or intraparticle diffusion. As they act in series, the slower of the two will be the rate determining step [29]. The possibility of the heavy metal ions to diffuse into the interior sites of the particles of adsorbent was tested with Weber-Morris equation [28] given as follows:

$$q = K_p t^{1/2} \quad (3)$$

Where, q is the amount of heavy metals adsorbed, K_p is the intraparticle diffusion rate constant and t is the time (agitation time) in minutes.

The rate constant K_p for intraparticle diffusion for various initial concentrations of heavy metal adsorption using groundnut shells and maize cob were determined from the slope of respective plots of amount of heavy metals adsorbed (q) vs square root of time (√t).

Figure 3: Plot of Elovich equation for the Adsorption of Pb (II) ion onto Groundnut shells and Maize cob.

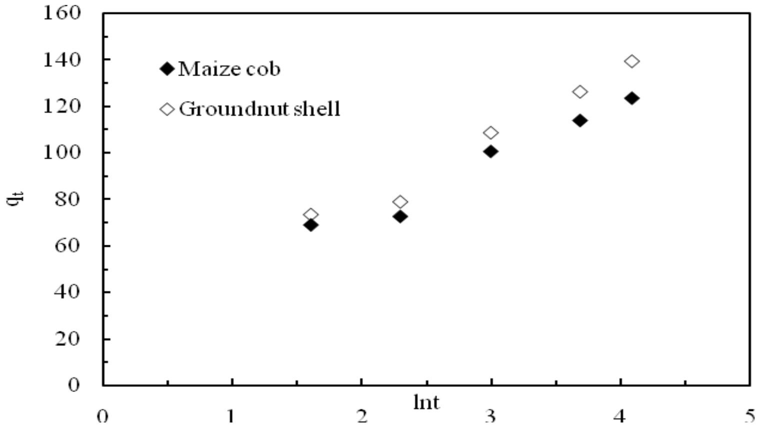


Figure 4: Plot of Elovich equation for the Adsorption of Zn (II) ion onto Groundnut shells and Maize cob.

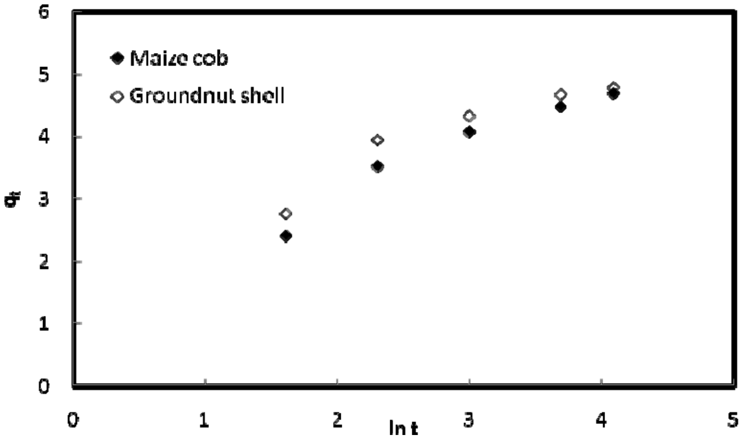


Figure 5: Plot of Intraparticle diffusion equation for the Adsorption of Pb (II) ion onto Groundnut shells and Maize cob

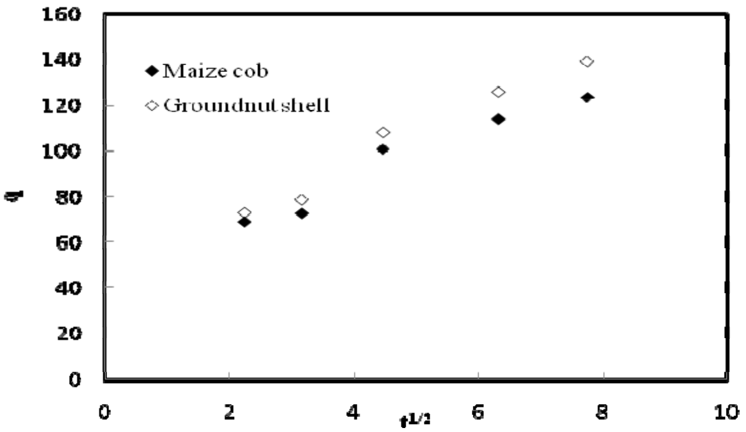


Figure 6: Plot of Intraparticle diffusion equation for the Adsorption of Zn (II) ion onto Groundnut shells and Maize cob.

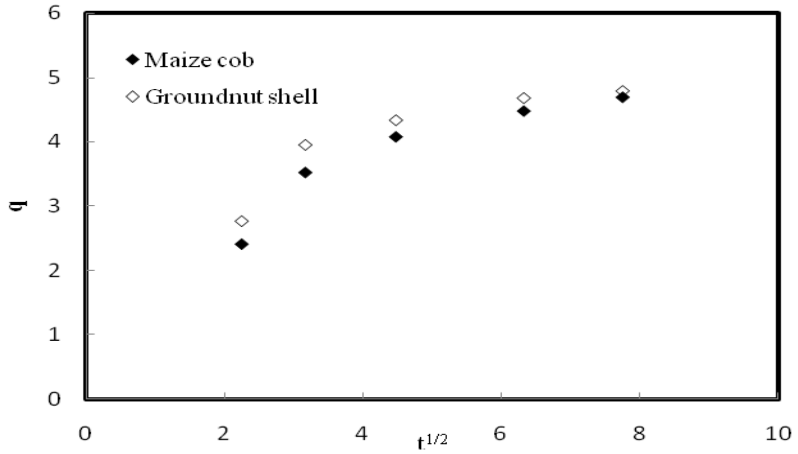


Table 4: Regression equations and R^2 values for the Elovich and Intraparticle equations for the Adsorption of Pb (II) and Zn (II) ions

Elovich equation		
Metal ions	**Groundnut shell**	**Maize cob**
Pb (II)	$y = 23.69x + 26.42$ $R^2 = 0.958$	$y = 28.12x + 22.55$ $R^2 = 0.968$
Zn (II)	$y = 0.765x + 1.860$ $R^2 = 0.891$	$y = 0.879x + 1.251$ $R^2 = 0.943$
Intraparticle diffusion equation		
Metal ions	**Groundnut shell**	**Maize cob**
Pb (II)	$y = 12.58x + 44.96$ $R^2 = 0.969$	$y = 10.53x + 45.56$ $R^2 = 0.949$
Zn (II)	$y = 0.319x + 2.581$ $R^2 = 0.776$	$y = 0.372x + 2.052$ $R^2 = 0.847$

Figures 3 and 4 shows a plot of the Elovich equation for the adsorption of Pb (II) and Zn (II) ions on Groundnut shell and Maize cob, while Figures 5 and 6 show a plot of the Intraparticle equation for the adsorption of Pb (II) and Zn (II) ions on Groundnut shell and Maize cob. The regression equations and R^2 values for the Elovich and Intraparticle equation plots are shown on table 4. A fairly linear relationship was obtained between metal ions adsorbed, q and ln t, q and $t^{1/2}$ over the whole adsorption period with high R^2 values that show a good fit. From the R^2 values on table 4, we discovered that the Elovich equation was more successful in describing the kinetics of heavy metal sorption. Table 4 also shows higher R^2 values for adsorption using maize cobs than groundnut shell hence maize cob is a better adsorbent for kinetic studies.

Conclusion

The results of this work has shown that agricultural by-products which are inexpensive and effective metal ion adsorbents can be carbonized, activated and used effectively as prepared from groundnut shell and maize cob for the removal of toxic lead and zinc and possibly other metals from their sources. It was also possible to use Elovich and Intraparticle equations to model the adsorption process. The regression models that were generated from this equation were used as predictive models for adsorption of these heavy metals onto groundnut shell and maize cob. Also, the results of this study could serve as parameters for designs of treatment plants for the treatment of heavy metal bearing effluents using groundnut shell, maize cob or even any other agricultural by-products as adsorbents. The capacity of the prepared activated carbons and their efficiency for the removal of Pb(II) ion and Zn(II) ion from aqueous solution was found to be adequate and therefore suitable for use in place of the costly commercial activated carbons.

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