



Zn(II) adsorption from synthetic solution and kaolin wastewater onto vermicompost

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ABSTRACT

The adsorption of Zn(II) from both synthetic solution and kaolin industry wastewater by cattle manure vermicompost was studied. The adsorption process was dependent on the various operating variables, viz., solution pH, particle size of the vermicompost, mass of vermicompost/volume of the Zn(II) solution ratio, contact time and temperature. The optimum conditions for Zn adsorption were pH 6.0, particle size of $\leq 250 \mu\text{m}$, 1 g per 10 mL adsorbent dose, contact time of 4 h and temperature of 25°C . Langmuir and Freundlich adsorption isotherms fit well in the experimental data and their constants were evaluated, with R^2 values from 0.95 to 0.99. In synthetic solution, the maximum adsorption capacity of the vermicompost for Zn^{2+} ions was 20.48 mg g^{-1} at 25°C when the vermicompost dose was $1 \text{ g } 10 \text{ mL}^{-1}$ and the initial adjusted pH was 2. The batch adsorption studies of Zn(II) on vermicompost using kaolin wastewater have shown the maximum adsorption capacity was 2.49 mg g^{-1} at pH 2 (natural pH of the wastewater). The small values of the constant related to the energy of adsorption (from 0.07 to 0.163 L mg^{-1}) indicated that Zn^{2+} ions were binded strongly to vermicompost. The values of the separation factor, R_L , which has been used to predict affinity between adsorbate and adsorbent were between 0 and 1, indicating that sorption was very favorable for Zn(II) in synthetic solution and kaolin wastewater. The thermodynamic parameter, the Gibbs free energy, was calculated for each system and the negative values obtained confirm that the adsorption processes are spontaneous. The ΔG° values were $-19.656 \text{ kJ mol}^{-1}$ and $-16.849 \text{ kJ mol}^{-1}$ for Zn(II) adsorption on vermicompost in synthetic solution at pH 6 and 2, respectively, and $-13.275 \text{ kJ mol}^{-1}$ in kaolin wastewater at pH 2.

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1. Introduction

One important source of heavy metal pollution in surface waters is the direct or indirect discharge of wastewater from kaolin industry. Brazil is the second largest kaolin producer in the world (reserves of 14%). The mineral, $\text{Al}_4(\text{OH})_8[\text{SiO}_4]_3$, is found in natural deposits of varied composition and among the uses of kaolin, stands out the utilisation as filler and coating in paper manufacturing, enhancing its density, brightness and smoothness [1].

Some contaminants of kaolin reduce its industrial application [2]. In order to improve kaolin quality it is necessary to remove contaminants such as iron oxides. The reduction of Fe(III) to Fe(II) is accomplished by using metallic Zn. Thus, Zn, Fe and Al are contaminants usually found in effluents from kaolin processing plants.

Vermicompost from different sources such as municipal solid waste [3], pig manure [4] and decomposed pods of green gram [5] promote plant growth. Vermicompost can also be obtained when cattle manure, together with soil, are used for earthworm diet. Earthworm species such as *Eisenia foetida* (Red of California), *Lumbricus terrestris* and *Lumbricus rubellus* have been successfully used in composting processes [6].

Substitution of conventional systems of effluent treatment is of particular importance. Recovered minerals or raw materials which have been processed into materials do not necessarily have to go to waste. Apart from critical environmental arguments, economic aspects speak for the reduction of metal wastes. In this context precipitation with lime (hydrous CaO), which is traditionally used for the removal of heavy metals from liquid effluents, can lead to an even more serious impact on heavy metal remobilization. The waste obtained is stored in large trenches and is not commonly reused. The stockpiling of such kind of residues on land may result in insidious leaching of metal by local acidic soils. In addition, the high price of lime compared to vermicompost (1 ton of lime costs about 4.3 times that of vermicompost) in Brazil, is other limiting factor.

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The processing of wastewater should avoid the metal enrichment of river water, leading to a better environmental condition. The kaolin processing plants contribute to water contamination by heavy metals. Thus, it is necessary to develop low cost processes of effluent treatment.

At present there are only a few studies regarding the treatment of wastewaters containing heavy metals by vermicompost. The efficiency of this substrate for removing Cu, Cr, Ni, Zn and Cd from synthetic solutions and electroplating wastes under laboratory conditions were studied by Jordão et al. [7]. The authors found that metal concentrations in the purified effluents were below the maximum values established for waste discharges into rivers by the Brazilian Environmental Standards. They also reported that the vermicompost residues obtained from the metal retention process could be applied as a fertilizer to agricultural lands.

In a similar work, Jordão et al. [7] reported that Cu, Zn and Ni retention by cattle manure vermicompost from electroplating wastes were close to 100%. They also reported that it was not necessary to correct the effluent pH during the treatment process to reach the levels recommended by Brazilian legislation for discharge into water courses. The potential application of vermicompost to adsorb Cd from both synthetic solution and mineral water was evaluated by Pereira and Arruda [8]. They found that vermicompost presented an expressive Cd adsorption capacity (38.6 mg g^{-1}) when compared with other adsorbents.

Adsorption processes are found to be highly effective, cheap and easy to adapt. Langmuir and Freundlich isotherms have been commonly used to model data obtained in wastewater adsorption treatment systems [9,10]. Thus, with the purpose of establishing a preliminary report for the removal of Zn(II) from synthetic solution and kaolin wastewaters, experimental equilibrium data obtained for Zn adsorption on cattle manure vermicompost has been analyzed by the Langmuir and Freundlich isotherm equations. Experiments including the effects of particle size, mass of vermicompost/volume of the synthetic Zn(II) solution ratio, contact time, pH and temperature have also been conducted before the experiment to optimize the maximum adsorption capacity.

2. Materials and methods

2.1. Sample collection and handling

Commercial samples of vermicompost of cattle manure (Super Húmus), which was produced in the Vista Alegre farm at the city of Ubá (State of Minas Gerais) was used as the adsorptive material. The raw vermicompost was air-dried for 72 h and used in the characterization experiments. For the adsorption experiments, the raw vermicompost was dried at 70°C for 4 h.

Kaolin wastewater was obtained from a factory located in Mar de Espanha at the Minas Gerais State, where this mineral is mined and kaolin processing is conducted. The sample was collected in a plastic bottle that it was previously soaked in diluted HNO_3 , rinsed with deionized water and wastewater before filling and refrigerated at 4°C .

Two additional samples of treated kaolin wastes from the factory were also collected in plastic bottles and included a limed waste sample and a decanted limed waste sample. However, these samples were not used in this work for Zn adsorption studies, since their Zn concentrations (0.44 and 0.55 mg L^{-1} , respectively) were below the maximum limit allowed by the Brazilian Environmental Standards [11] for industrial effluent discharges into water systems ($<5 \text{ mg L}^{-1}$). These samples had the pH values of 11.24 and 12.24.

2.2. Vermicompost and kaolin wastewater characterization

The vermicompost pH was measured in deionizer water (solid/solution ratio of 1:2.5) using a pH meter. Moisture content of the vermicompost was determined by the percentage loss in weight after drying the sample at 60°C and at 110°C for 24 h; organic matter content was measured by ignition in a furnace at 550°C for 24 h and ash content after heating at 800°C for 2 h [12]. The C and H contents were measured with an infrared detector and the N content was measured with a thermal conductivity detector.

The carboxylic group content was determined in approximately 0.1 g of vermicompost. This amount was added to 10 mL of $0.5 \text{ mol L}^{-1} \text{ Ca(OAc)}_2$ solution. After agitation of the mixture for 24 h, the carboxylic groups were determined by titration with a $0.1 \text{ mol L}^{-1} \text{ NaOH}$ solution to pH 9.8 [13].

A non-linear regression equation was adjusted to the titration data to differentiate the acid groups. For this purpose, 1 g of the vermicompost, previously sieved to a particle size of less than 0.177 mm, was titrated potentiometrically with NaOH solution and the pK_a values determined.

The particle size distribution was evaluated by a pipette method, using $1 \text{ mol L}^{-1} \text{ NaOH}$ solution as a dispersant agent [14].

The total concentrations of Cu, Ni, Zn, Mn, Fe, Pb, Cd, Ca, Mg, Na and K were determined in vermicompost using 0.5 g portions of air-dried samples. They were digested individually at 200°C with 5 mL of 65% (w/v) HNO_3 . A 5 mL aliquot of concentrated HClO_4 (70% w/v) as well as a 5 mL aliquot of HF (40% w/v) were added and the mixtures re-evaporated to near dryness. Finally, a 5 mL aliquot of 65% (w/v) HNO_3 was added. The mixture was re-evaporated to near dryness and diluted with deionized water to 25 mL [7]. The total metal concentrations in the resultant solutions were then measured by atomic absorption spectrophotometry (AAS).

The kaolin wastewater pH was measured by immersing a volume of 50 mL of the sample into the instrument cell. The values of the pH of limed waste sample and decanted limed waste sample were obtained in the resultant solutions after filtration. The concentrations of Fe, Mn, Cd, Cu, Cr, Ni, Zn, Al, Ca, Mg and Pb were determined in the wastewater after filtration by inductively coupled plasma optical emission spectrometer (ICP-OES).

2.3. Metal adsorption studies

2.3.1. Preliminary experiments for Zn(II) adsorption optimization

All the parameters such as particle size range, mass of vermicompost/volume of the synthetic Zn(II) solution ratio, contact time, adsorption pH and temperature were optimized for the maximum adsorption capacity of the vermicompost. Batch adsorption experiments were carried out in 50 mL centrifuge tubes containing vermicompost sample in which it was added 10 mL of $150 \text{ mg L}^{-1} \text{ ZnCl}_2$ (in $0.1 \text{ mol L}^{-1} \text{ KCl}$ in order to equalize ionic strength). The suspensions were mechanically stirred at 120 rpm, centrifuged and the Zn concentrations determined in the solutions. For optimization, particle size was varied between ≤ 105 and $\leq 2000 \mu\text{m}$ and mass of vermicompost/volume of the Zn(II) solution ratio varied between 0.25 g and 1 g 10 mL. The shaken time was examined in the range 1–28 h and solution pH in the range of 3.0–7.0. Temperature was evaluated in the range of 10 ± 2 – $40 \pm 2^\circ\text{C}$.

Each preliminary experiment was replicated three times and the Zn concentrations were determined by AAS. The adsorption of Zn(II) was calculated as the difference between that added and that in the supernatant.

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