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Research Article

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Biosorption of zinc (II) ions from aqueous solution by *Androgaphis paniculata* leaves powder on batch method

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ABSTRACT

The research about biosorption of Zn(II) by Androgaphis Paniculata leaves has been investigated through batch experiment. Based on the experiment, optimum condition for adsorption Zn (II) ion was pH 6, biosorbent dosage at 0.25 g, initial concentration of metal ion solution 2400 mg/L and contact time 15 minutes. The biosorption data were fitted to Freundlich isotherm with R^2 *0.9560. The maximum biosorption capacities were estimated from Freundlich isotherm model to be 86.96 mg/g. Characterization of Androgaphis paniculata was is evaluated using Fourier Transform Infra Red (FT-IR) spectroscopy and active functional group estimated were O-H group, C-H aliphatic, C=O, and C-C aliphatic. The measurement of metal ion concentration conducted by Atomic Absorption Spectroscopy (AAS) and surface analysis before and after adsorption conducted using Scanning Electron Microscopy (SEM). This study showed that Androgaphis Paniculata leaves powder could be used as an efficient adsorbent to remove Zn (II) metal ions.*

Keywords: Biosorption, *Androgaphis paniculata*, Freundlich isotherm, Biosorption capacity

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INTRODUCTION

Presently, the development of chemical industries to meet human needs is very progressing, especially in electroplating industry. Modern human lives cannot be separated from electro planting consumer products such as household products, automotive products, etc. Therefore, these products are being used widely in daily lives. Electro planting activity not only produces useful products but also waste products that affect human lives. Liquid waste containing heavy metal will be causing danger to the environment. The heavy metal will accumulate and persist, so that it affects human health [1]. One of the heavy metals that caused danger to the ecosystems and environmental health is Zn (II) ion. Zn (II) ion in human body is not biodegradable, so it can accumulate in human organs [2].

Mythill, et al study, in "Analysis Heavy Metal Content in *Androgaphis Paniculta*" (2011) Journal used *Androgaphis Paniculata* as a biomass. The biomass then mixed into 1000 mg/L heavy metals (Fe, Cr, Cu, Cd) and measured by Atomic Adsorption Spectrophotometry (AAS) which showed significant contents of heavy metal Fe and Cr in the *Androgaphis Paniculata* leaves. Therefore, author will study the usage of *Androgaphis Paniculata* leaves to overcome Zn (II) ion toxicity and DNA damage in mouse [3].

The objectives of this study are characterization of *Androgaphis Paniculata* leaves as a biomaterial to absorb Zn (II) ion, studying lethal dose of Zn (II) ion injected to trial mouse and the damage of trial mouse kidney organ by the DNA damaged caused. The characterization includes pH influence, Zn (II) ion concentration, *Androgaphis Paniculata* leaves powder weight, the contact time between *Androgaphis Paniculata* leaves powder and Zn (II) ion.

EXPERIMENTAL SECTION

Equipments

Volumetric flask, measuring pipette, erlenmeyer (Iwaki PYREX), plastic vial bottle, plastic funnel, and filter paper. Electronic equipments: analytics weighter (Kern & Sohn GmbH), Rotary Shaker (Edmund Buhler 7400 tubingen), pH meter (LovibondSenso Direct), Atomic Adsorption Spectroscopy (Varian Spektro AA 240 Spectrometer), Fourier Transform Infra Red (Thermo Scientific Nicolet IS10 using KBr pellets), Scanning Electron Microscope (Hitachi Model S-3400N).

Materials

Androgaphis Paniculata, $(CH_3COO)_2Cd.2H_2O$ crystal (Merck), NaOH crystal (Merck), HNO_3 p.a (Merck), CH_3COONa crystal (Merck), CH_3COOH (Merck), NH_4OH (Merck), NH_4Clcystal (Merck), H_3PO_4p.a (Merck), KH_2PO_4crystal (Merck), K_2HPO_4crystal (Merck) and Aquadest.

Method

The method used in this study is Batch system.

Early treatment of sample

Androgaphis Paniculata leaves sample collected in Medan, then washed in water, and dried in Environment Analytic Chemical Laboratory, Andalas University, Padang. The dried sample then was mashed by blender.

Biosorbent Activation

20 g of mashed *Androgaphis Paniculata* sample then collected in beaker glass, and was added by 80 mL of HNO_3 0.01 M for 2 hours. After that, washed with aquadest until neutral pH reached, filtered, and then dried so the sample is ready for used in the determination of metal ion adsorption optimum condition.

Preparing Zn 100 mg/L Solution

$ZnSO_4$ 1000 mg/L solution was obtained by dissolving 4.3950 g of $ZnSO4.7H_2O$ until it reached 1 liter solution with aquadest as the solvent.

Determining Optimum Conditions for Adsorption

Effect of pH solution towards metal ion adsorption

10 mL of Zn 30 mg/L solution with pH vary from 3, 4, 5, 6, 7, dan 8 were put into 25 mL erlenmeyer. Then 0.25 g biosorbent *Androgaphis Paniculata* leaves powder added, closed with aluminium voil and shaked for 15 minutes at 100 rpm. Solution filtered with filter paper and the metal ion concentration of the filtratedetermined by AAS. Calculate the adsorption capacity (Q) for each pH and determine the optimum pH measurement.

Effect of Zn (II) metal concentration towards metal ion adsorption

Each 10 mL of Zn solution with different concentration of 30, 60, 90 and 120 mg/L were put into 25 mL Erlenmeyer. Then 0.25 g biosorbent *Androgaphis Paniculata* leaves powder added, closed with aluminium voil and shaked for 15 minutes at 100 rpm.. Solution filtered and the metal ion concentration of the filtrate determined by AAS. Calculate the adsorption capacity (Q) for each Zn (II) metal concentration and determine the optimum Zn (II) metal concentration measurement.

Effect of biosorbent mass towards metal ion adsorption

10 mL of Zn (II) solution with the optimum concentration and pH were put into 4 pieces of Erlenmeyer, then added by biosorbent *Androgaphis Paniculata* leaves powder weighting 0.1; 0.25; 0.5 and 1.0 g, shaked for 15 minutes at 100 rpm. Solution was filtered and the biosorption result of the filtrate measured with AAS. Calculate the adsorption capacity (Q) for each biosorbent mass and determine the optimum biosorbent mass measurement.

Effect of contact time towards metal ion adsorption
10 mL of Zn (II) solution with the optimum concentration and pH were put into 4 pieces of Erlenmeyer, then added by biosorbent *Androgaphis Paniculata* leaves powder weighting the optimum mass and shaked for 15, 45, 90 dan 120 minutes at 100 rpm. Solution was filtered and the metal ion concentration of the filtrate measured with AAS. Calculate the adsorption capacity (Q) for each contact time and determine the optimum contact time measurement.

Adsorption Isotherm Analysis
Data from the study were determined whether Langmuir Isotherm or Freundlich Isotherm.

FTIR Analysis
Biosorbent FTIR analysis was done before and after metal ion adsorption. 0.1 g of *Androgaphis Paniculata* leaves powder was mashed with KBr, shaped into thin disc or pellets. Sample was analyzed with FTIR to determine the functional group of *Androgaphis Paniculata* leaves powder.

SEM Analysis
Androgaphis Paniculata leaves powder was analyzed by SEM before and after metal ion adsorption to determine the biosorbent surface morphology.

RESULTS AND DISCUSSION

Effect of pH solution
pH is an important parameter in the adsorption process of Zn(II) ions by biomaterials. The effect of pH solution was studied in range of 3 to 8. Fig.1 shows that at pH 3 the adsorption capacity of *Androgaphis Paniculata* is low due to the presence of H3O+ ions in solution. At higher pH solution the competition for metal ion in active site with H3O+ decreases, so the adsorption capacity increases [4]. Adsorption capacity of Zn(II) Ions increased at pH 5 to pH 6. The optimum adsorption capacity of Zn(II) ions occur at pH 6 for *Androgaphis Paniculata* leaves. At pH 6 to 7 the adsorption capacity of Zn(II) ions were reduced.

Effect of Initial Metal Concentration
The initial concentration of metal ions was studied to determine the ability of the active site of biosorbent to bind metal ions [5]. The amount of metal ions adsorbed by *Androgaphis Paniculata* leaves increased with the increasing of metal concentrations. However, capacity is reduced when high concentrations is reached due to the active site that already bonded by metal ions, this causing the saturation and there are no more active site of biosorbent remains to bind other metal ions. In this case the equilibrium between the metal ions adsorbed by biosorbent achieved. Fig.2 shows that the optimum adsorption capacity of Zn(II) ions were found when the concentration is 2400 mg/L with *Androgaphis Paniculata* leaves.

Effect of biosorbent mass towards metal ion adsorption
Fig. 3 shows the optimum biosorbent mass for bio material was found to be 0.1 g. The amount of adsorbent used represents the potential of biosorbent in removing ions on the initial concentration given [6]. The metal ion adsorption capacity is inversely proportional to the mass of biosorbent used. Increased of biosorbent mass resulted the decline in metal ion adsorption capacity. A specific area can be defined as a part of total area available for biosorption [7].

Effect of Contact Time
Fig. 4 shows that the contact time for the adsorption process of Zn(II) ions reach the optimum value in 15 minutes contact time with *Androgaphis Paniculata* Leaves. It decreases after 25 minutes, then it increases again until 120 minutes. This decreasing is caused Zn(II) ions which has been bound to the active site to become loose back and caused the adsorption capacity to be decreased.

Adsorption isotherms
Equilibrium relationship between adsorbent and adsorbate can be explained by adsorption isotherms [8]. Determination of equilibrium parameters provides important information that allows advance adsorption design of the system [9]. Biosorption isotherms are characterized by certain parameters, which explain the nature of the surface and biosorbent affinity. It is also used to compare the biosorptive capacity and biosorbent for different pollutants [10]. Data trial uptake of Zn(II) ions for biomaterial was analyzed by Langmuir and Freundlich isotherm

models. Langmuir isotherm models assume a surface with homogeneous binding sites, equivalent energy adsorption, and no interaction between species adsorbed [11]. Freundlich isotherm models assume the adsorption occur on heterogeneous surfaces with interaction between molecules adsorbed, the application of Freundlich equation also indicates that the energy adsorption decreases exponentially at the completion of the adsorption center of adsorbent [12]. Fig.5 shows that R2 value for Zn(II) ions is 0.9560 with *Androgaphis Paniculata* Leaves and follows Freundlich adsorption models, while Fig.6 shows that the isotherm models of Zn(II) ions gave the determination coefficient value (R2) of 0.9398 that follows Langmuir adsorption models.

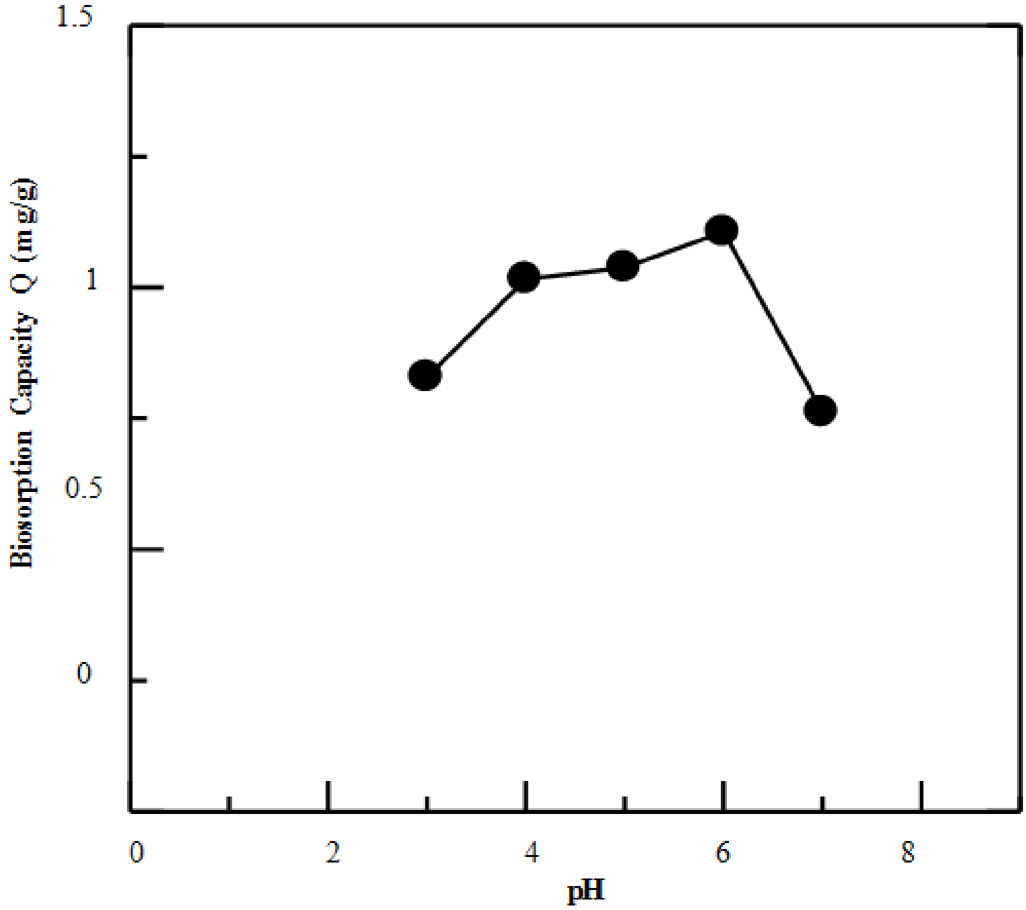

Figure 1. Effect of pH towards Zn (II) metal biosorption process by using *Andrographis Paniculata* leaves powder. The optimum pH for 30 mg/L Zn (II) metal biosorption process by using *Andrographis Paniculata* leaves powder was at 6

Analysis of functional groups by FTIR

FTIR analysis of *Androgaphis Paniculata* leaves was shown in Fig.7. The analysis showed ions removal mechanism in adsorbent surface, because it provides information about the functional groups present in the structure of the adsorbent [6]. A wavelength of 3404.97 cm-1, shows the presence of hydroxyl (-OH) stretching groups caused by inter-molecular hydrogen bonding compounds such as phenols, alcohols and carboxylic acids. A wavelength of 1647.96 cm-1 is a spectrum of C=O groups in amide. At a wavelength of 1323.86 cm-1 peak of the cluster C-N amine and C-O stretching of alcohol groups were found. The wavelength of 1053.70 cm-1 indicates the C-N bond stretching. Wave length of 3404.97 cm-1 becomes 3023.36 cm-1. This FTIR spectra can provide information that hydroxyl was involved during the adsorption process of Zn(II) ions.

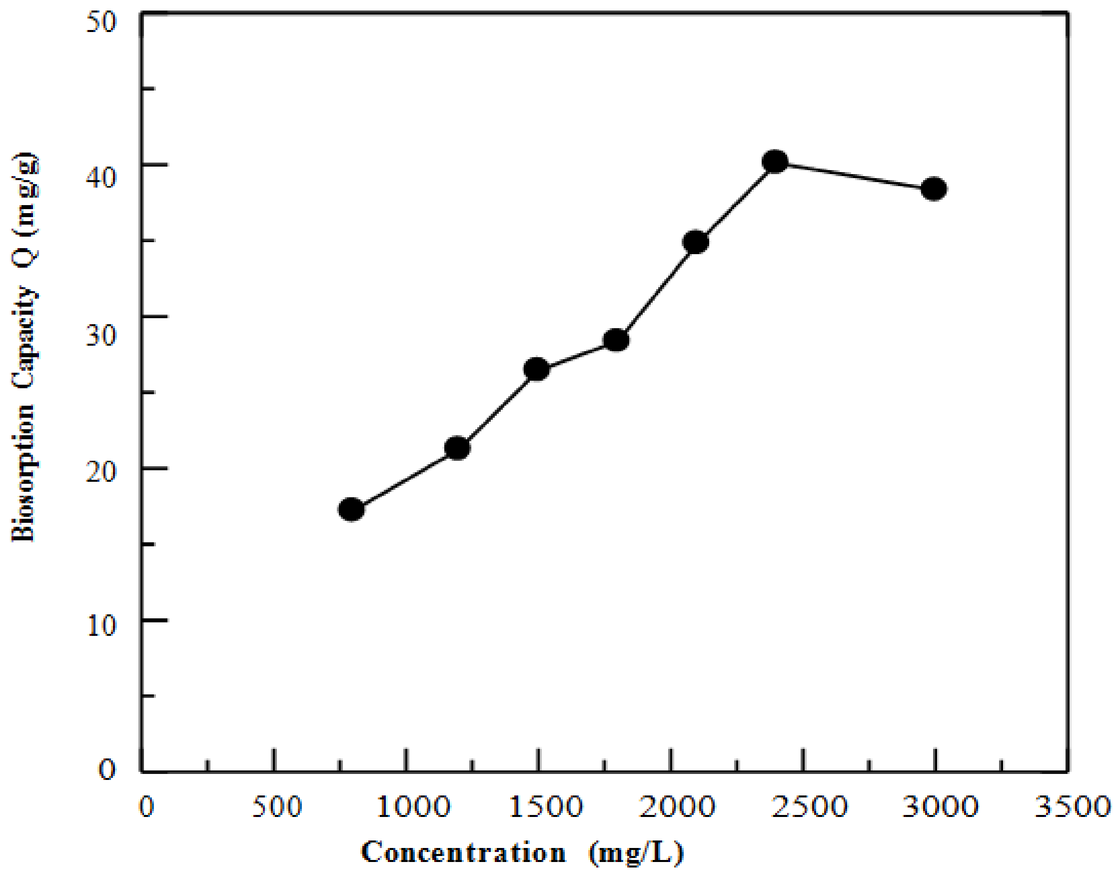

Figure 2. Effect of concentration towards Zn (II) metal biosorption process by using Andrographis Paniculata leaves powder. The optimum Zn (II) metal concentration on biosorption process by using Andrographis Paniculata leaves powder was 2400 mg/L

Analysis of SEM

Characterization using SEM was done to see the morphology structure of adsorbent material. Characterization is done at a magnification of 1000x. Fig. 8 shows irregular structure of *Androgaphis Paniculata* leaves before Zn(II) adsorption and the presence of empty pores. These pores will facilitate the adsorption place of metal ions. Fig. 9 shows that the empty pores has been filled with metal ions after the process Zn(II) adsorption.

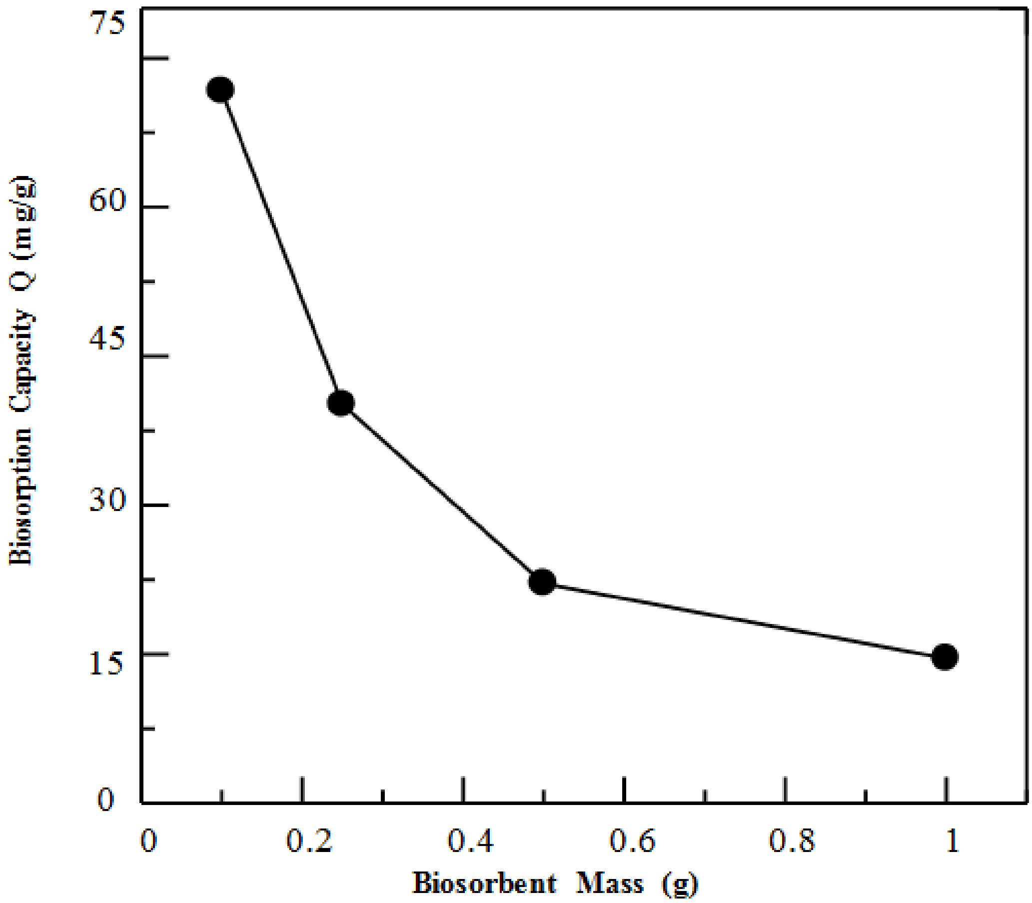

Figure 3. Effect of biosorbent mass towards Zn (II) metal biosorption process by using *Andrographis Paniculata* leaves powder. The optimum biosorbent mass on biosorption process of Zn (II) metal by using *Andrographis Paniculata* leaves powder was 0.25 g.

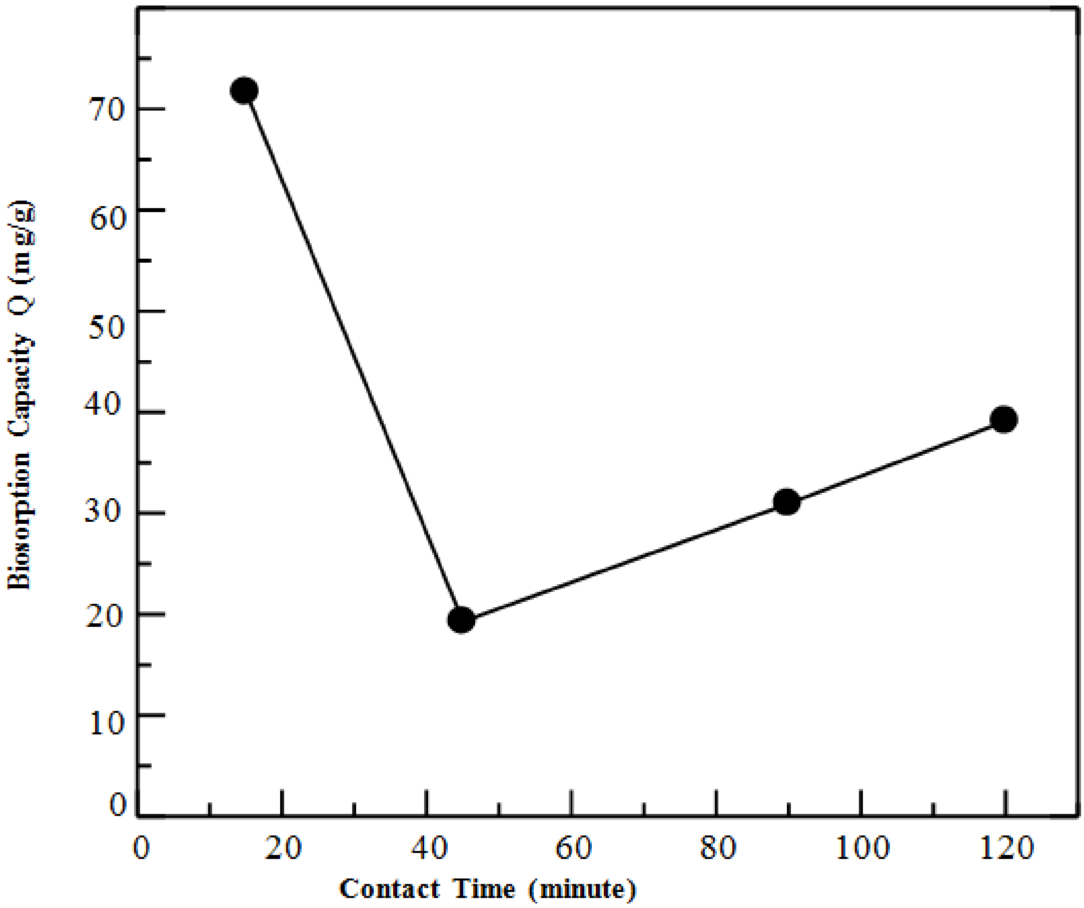

Figure 4. Effect of Zn (II) metal biosorption process contact time by using *Andrographis Paniculata* leaves powder. The optimum contact time on biosorption process of Zn (II) metal by using *Andrographis Paniculata* leaves powder was 15 minutes

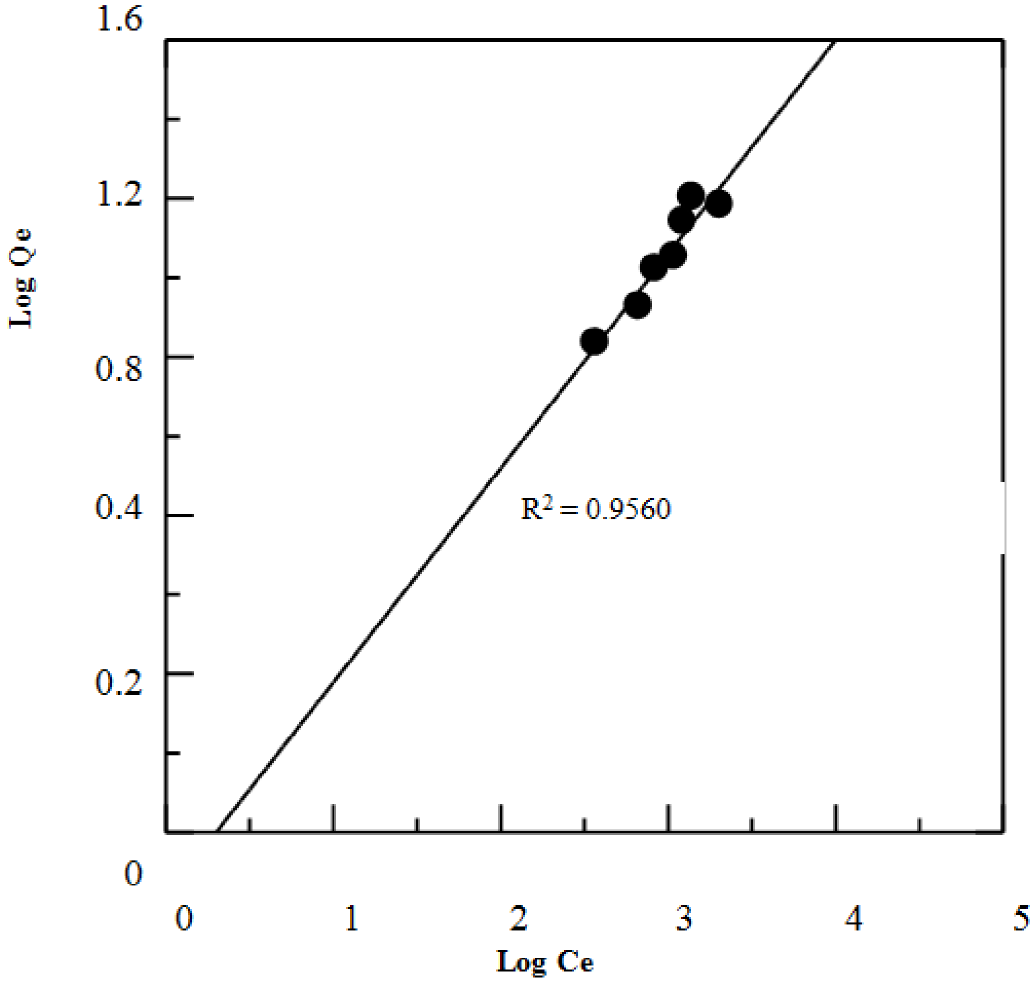

Figure 5. Freundlich Isotherm, Plot C_e vs C_e/Q_e obtained regression equation of: y = 17.16 + 0.0166 x

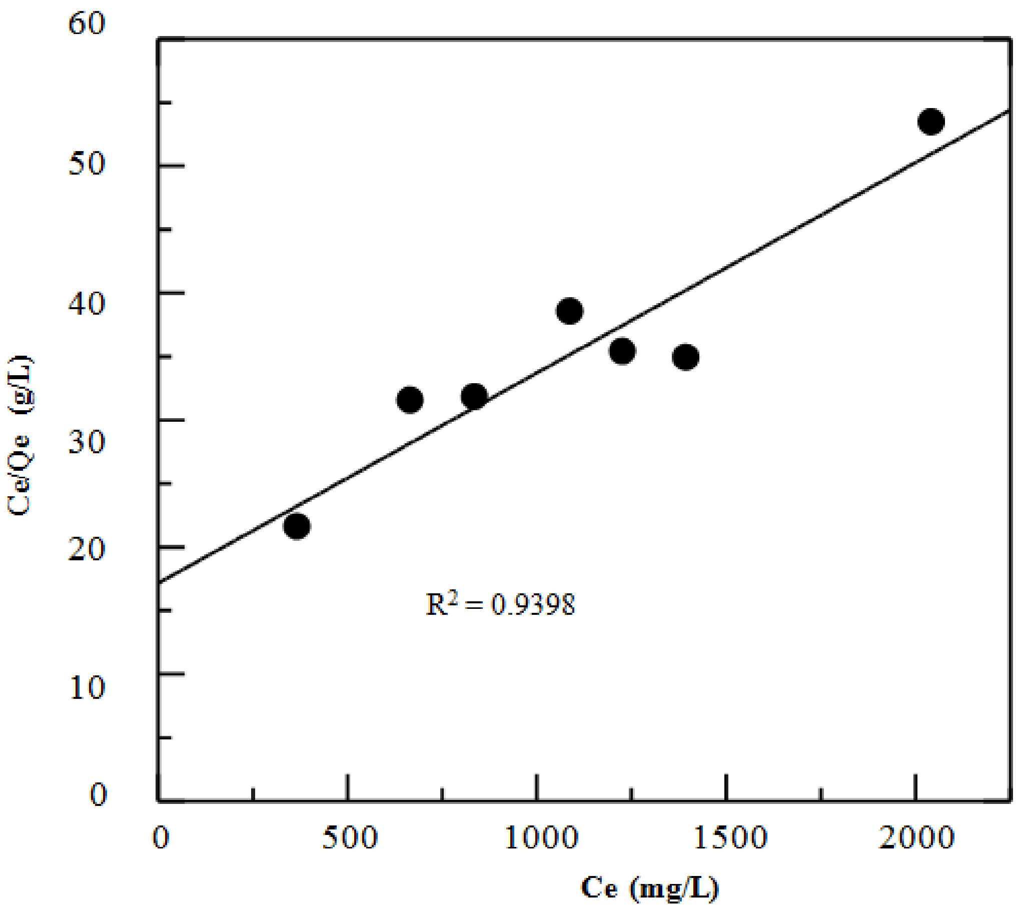

Figure 6. Langmuir Isotherm. Plot $C_e$vsC_e/Q_e obtained regression equation of: y = 17,16 + 0,0166x

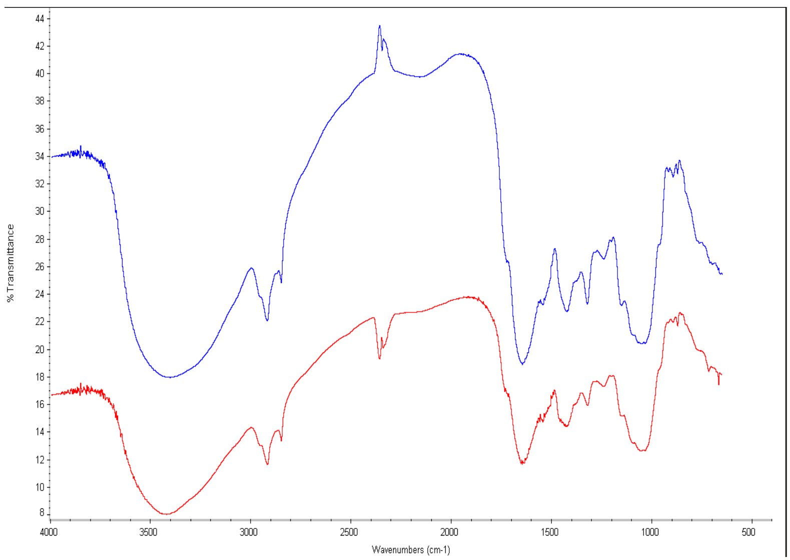

Figure 7. FTIR Spectrum of *Andrographis Paniculata* leaves powder

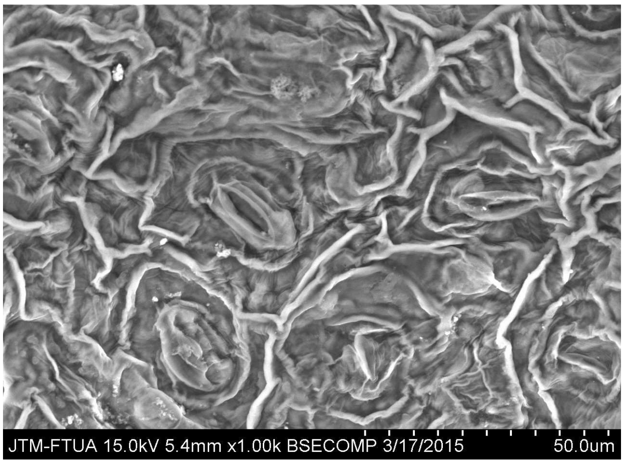

Figure 8. Scanning Electron Microscope (SEM) of *Andrographis Paniculata* leaves powder before Zn(II) ion adsorption Magnification 1.000 times

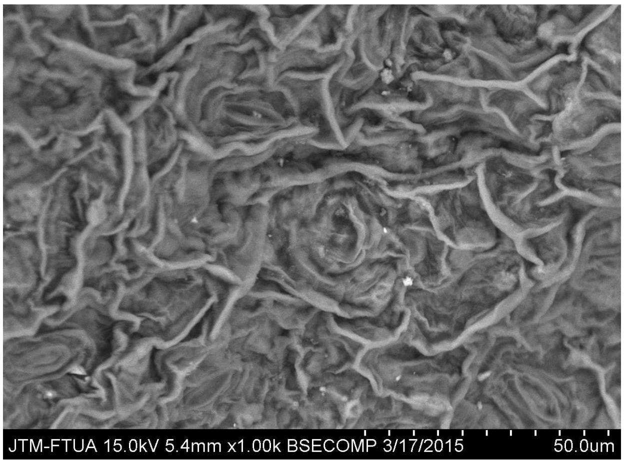

Figure 9. Scanning Electron Microscope (SEM) of *Andrographis Paniculata* leaves powder after Zn(II) ion adsorption Magnification 1.000 times

CONCLUSION

Based on this research, it can be concluded that the herbal *Androgaphis Paniculata* leaves can be used in Zn(II) adsorption. It is hoped that this herbal has a potential adsorption towards Zn(II) ions on animal organs.

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