



Research Article

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**Adsorption profile of Cu (II) using Soursop (*Annona muricata* L) leaves powder as biosorbent**

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**ABSTRACT**

Biosorption of Cu (II) ions from aqueous solutions onto soursop (*Annona muricata* L) leaves powder was investigated in a batch system. Several variables such as effect of pH, contact time, concentration of metal ions and biosorbent dosage have been investigated. The result showed that the adsorption process was strongly dependent with pH and initial concentration of Cu (II). The optimum condition was achieved for adsorption on pH 7 and initial concentration of Cu (II) was 500 mg/L with sorption capacity 6.14 mg/g. The optimum adsorption was also achieved with biosorbent dosage 0.25 g and contact time 90 minutes. The experimental equilibrium biosorption data were analyzed by Langmuir isotherm model with  $R^2 = 0.9689$ . Furthermore, the biosorbent was characterized by Fourier Transform Infra Red (FT-IR) spectroscopy to identify the various functional groups contributing in the sorption process. Scanning Electron Microscope (SEM) was used for surface analysis before and after adsorption process.

**Keywords:** Biosorption, Cu (II), *Annona muricata* L, Langmuir isotherm

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**INTRODUCTION**

Industrial wastewater often characterized by the presence of heavy metal content in the effluent, so it is requiring treatment prior to disposal in order to avoid water pollution. Various industries contain heavy metals in their aqueous waste streams, such as mining, metal plating, batteries, wire drawing, circuit board manufacturing, even agricultural source where fertilizer and fungicidal sprays are intensively used [1]. Examples of heavy metals released include nickel, lead, copper, zinc, cadmium and aluminum. These metals are toxic and might cause environmental pollution, and also could affected the human health as the ions are water soluble and could absorbed into human body [2]. Copper is a metal that extensively used in industries and is an essential element for human health. Copper is potentially toxic and exposure to copper can lead to liver damage causing gastrointestinal problems, Wilson Disease and Insomnia [3]. Copper initially depositing in the liver and interfere the liver's ability to detoxify elevated level of copper in the body thus adversely affect nervous system, reproductive system, connective tissue, adrenal function, learning ability of the new born, etc [4].

Because the various adverse effect of heavy metal contamination, the government may consider it necessary to remove some pollutants from ecosystem, however some conventional technologies such as precipitation and sludge separation, chemical oxidation and reduction, ion exchange, reverse osmosis, electrochemical and evaporation, need

very high cost. In the last two decades, biosorption technology has drawn attention because its low cost, high efficiency and minimization production of secondary chemical and biological sludge [5].

Nowadays various agricultural byproducts has been used to remove heavy metals from waste water such as mangosteen fruit shell [6], orange fruit peel [7], cacao shell [8], etc. *Annona muricata* L commonly called sour is a small erect evergreen tropical fruit tree plant and its well known as medicine of painful foot and back, and their leaves has been used as hypertension medicine [9]. The aim of this study is to investigate the ability of *Annona muricata* L leaves as biosorbent for Cu (II) in aqueous solution. The effect of pH solution, contact time, adsorbent dose, and initial concentration also studied. The characterization of functional group of biosorbent was determined by FTIR, the surface of biosorbent was analyzed by SEM. The Freundlich and Langmuir isotherm also studied to quantify the adsorption equilibrium.

## EXPERIMENTAL SECTION

### Chemical and Apparatus

All reagents used are analytical grade and obtained from E. Merck (Darmstad, Germany). The apparatus used are analytical balance (Kern & Soho GmbH), Rotary Shaker (Edmund Buhler 7400 tubingen), pH meter (Lovibond Sensodirect), Atomic Absorption Spectroscopy (Varian Spektro AA 240 spectrometer), Fourier Transform Infra Red (Thermo Scientific Nicolet IA10 using KBr pellets), Scanning Electron Microscope (Hitachi Model S 3400 N).

### Preparation of Adsorbent

*Annona muricata* L was collected from home garden in Padang, Indonesia, washed with deionized water, air dried and then ground using crusher. About 20 g of *Annona muricata* L leaves was soaking into 80mL HNO<sub>3</sub> 0,01M, followed by washing thoroughly with deionized water then air dried. The biosorbent is ready to be used.

### Biosorption Studies

Adsorption experiment was conducted at various pH of the solution, contact time, various initial concentrations of Cu (II) and biosorbent dosage. Characterization of biosorbent was conducted using FTIR and SEM before and after Cu (II) uptake. To determine the amount of Cu (II) ion adsorbed by *Annona muricata* L leaves powder (qe), the formula used is:

$$q_e = \frac{C_o - C_e}{m} \times v \quad (1)$$

where as Co is the initial concentration of metal ions (mg/L), Ce is final concentration at equilibrium state (mg/L), m is biosorbent mass (g) and v is volume solution (L).

## RESULTS AND DISCUSSION

### Effect of pH on biosorption of Cu(II) ions

Effect of pH solution was studied in pH range 3-8 with 10 mL Cu (II) 30 mg/L, biosorbent dose 0.25 g and contact time 15 min with stirrer speed 100 rpm. The result shown in Fig.1

Fig.1 shows that adsorption capacity gradually increased from 1,097 mg/g to 1.182 mg/g with increase of pH and then decreased. The optimum pH is at pH 7. pH is one critical factors that influence adsorption of metal ions since it directly affects the metal solubility or the dissociation degree of functional groups on the surface of sorbent. In general, alkaline conditions shall decrease the metal adsorption rate since an increase of pH it means a lower amount of protons, causes the competition between of proton and heavy metal ions decreases [10]. However, at low pH the H<sup>+</sup> ions shall compete with metal ions for the exchange sites on the biosorbent, led to partially releasing the cations which resulted in very low metal ions uptake [11]. The similar result has been reported by several authors [10, 12, 13].

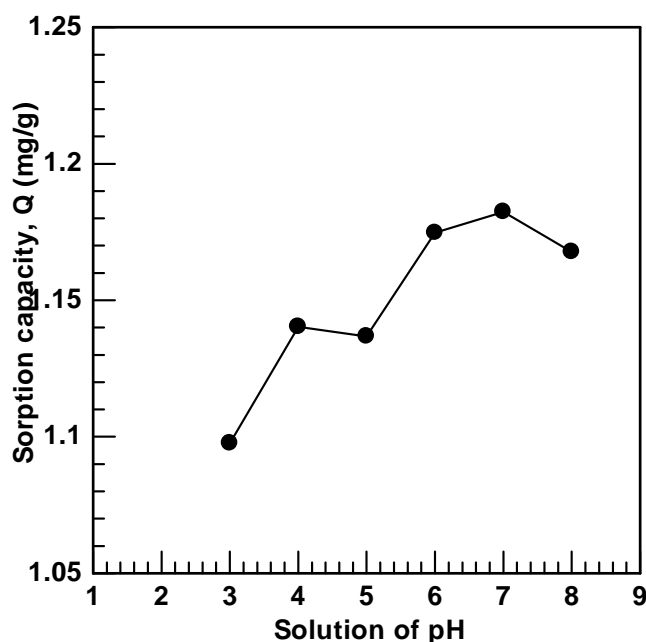


Figure 1. Effect of pH on Cu (II) bisorption by *Annona muricata* L; initial concentration of Cu (II) 30 mg/L; stirrer speed 100 rpm; contact time 15 min; biosorbent dose 0.25 g

#### Effect of initial concentration on biosorption of Cu (II) ion

Fig.2 illustrates the biosorption of Cu (II) ions by *Annona muricata* L leaves powder as a function of initial metal concentration.

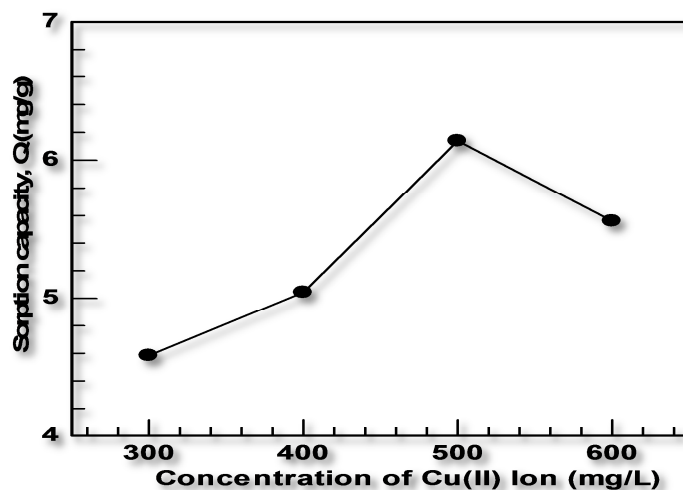


Figure 2. Effect of concentration on Cu (II) adsorption by *Annona muricata* L; pH 7; stirrer speed 100 rpm; contact time 15 min; biosorbent dose 0,25 g

Effect of initial concentration of Cu (II) was conducted at various concentrations ranging from 300 to 600 mg/L at pH 7. From Fig.2 the optimum adsorption capacity of *Annona muricata* L leaves powder was achieved at concentration Cu (II) 500 mg/L with value 6.14 mg/g. Biosorption capacity increased as the increase of initial concentration, and then decreased after achieve the equilibrium. Higher concentration led to a greater driving force at the liquid solid interface, which enhance the mass transfer [14].

#### Effect of biosorbent dosage on biosorption of Cu (II)

The amount of biosorbent that used in biosorption process is an important factor which determines the potential of biosorbent to remove the metal ions at a given initial concentration [15]. The dosage of *Annona muricata* L leaves powder used in this study varying from 0.1 – 1 g. The result was shown in Fig. 3.

As revealed from Fig.3 the sorption capacity was increased at low dose of biosorbent mass. The optimum sorption capacity for *Annona muricata* L leaves powder was 6.14 mg/g with biosorbent dose 0.25 g. The sorption capacity of biosorbent was decreased as the increase of biosorbent dosage. This could be explained that at a given concentration in suspension, the lower biosorbent concentration, the higher will be the metal and biosorbent ratio and the metal retained by a sorbent [10]. For the lower concentration of biosorbent, there is an increase in the uptake of metal ions [16].

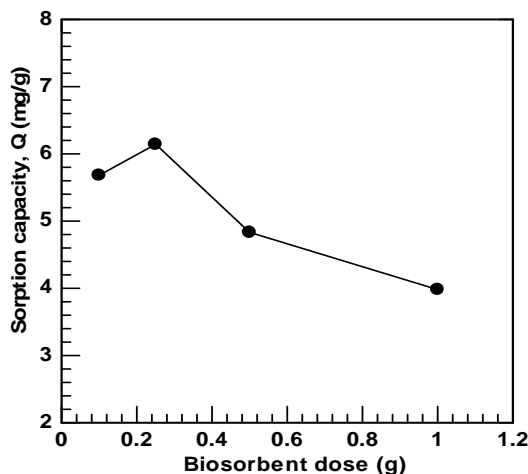


Figure 3. Effect of biosorbent dose on on Cu (II) biosorption; pH 7; stirrer speed 100 rpm; initial concentration 500 mg/L; contact time 15 min

#### Effect of contact time on adsorption of Cu (II) ion

The effect of contact time on biosorption of Cu (II) by *Annona muricata* L leaves powder was conducted with varying contact time from 20 to 120 min. as shown in Fig.4, the optimum sorption capacity of *Annona muricata* L leaves powder was achieved at contact time 90 min with value 10.032 mg/g.

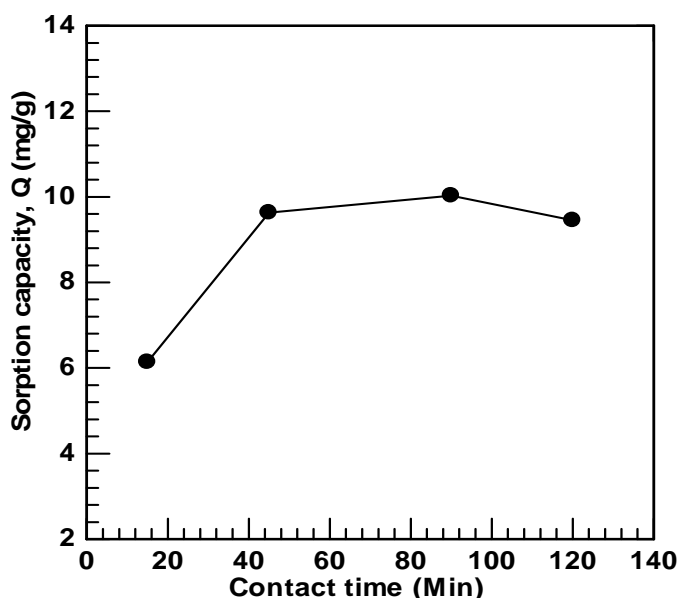


Figure 4. Effect of contact time on Cu (II) biosorption; pH 7, stirrer speed 100 rpm; initial concentration 500 mg/L; biosorbent dose 0.25 g

As revealed in Fig.4 the increased of sorption capacity was occur on the first 90 minutes contact time and then decreased. The contact time is an important parameter for an optimum removal of metal ions in aqueous solution. The increase of sorption of metal ions with the increase of contact time can be due to the decreased coefficient of mass transfer of the diffusion controlled reaction between the sorbent and the metal ions [17].

### FTIR Analysis

In order to estimate the functional groups which involved in biosorption process, FTIR analysis was conducted. The FTIR spectra of *Annona muricata* L leaves powder before and after biosorption of Cu (II) was shown in Fig.5.

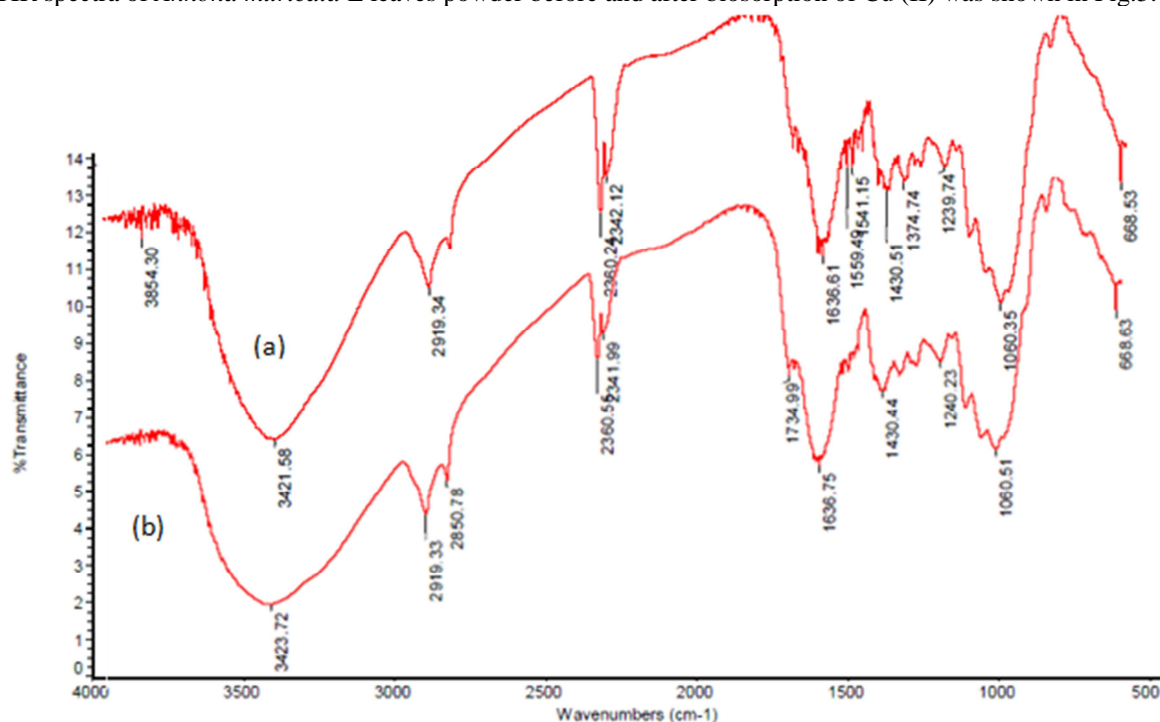


Figure 5. FTIR spectra of *Annona muricata* L: (a) before biosorption of Cu (II); (b) after biosorption of Cu (II)

As depicted in Fig.5 the broad and sharp peak at wavelength 3500-3200  $\text{cm}^{-1}$  indicates the O-H stretch from alcohols or phenol, the medium peak at 3300-2500  $\text{cm}^{-1}$  indicates the O-H stretch from carboxylic acid, and medium peak at 1650-1580  $\text{cm}^{-1}$  indicates the N-H bend. The shifted of the wavelength before and after biosorption indicates that there is a biosorption process occurs between *Annona muricata* L leaves powder and Cu (II) ion in aqueous solution.

### SEM analysis

SEM analysis was conducted to observe the morphology surface of *Annona muricata* L leaves powder before and after biosorption. The SEM image at Fig.6 with magnifying 1000x shows that *Annona muricata* L leaves have porous surface and heterogenous rough. It is indicate that *Annona muricata* L have a good possibilities as biosorbent.

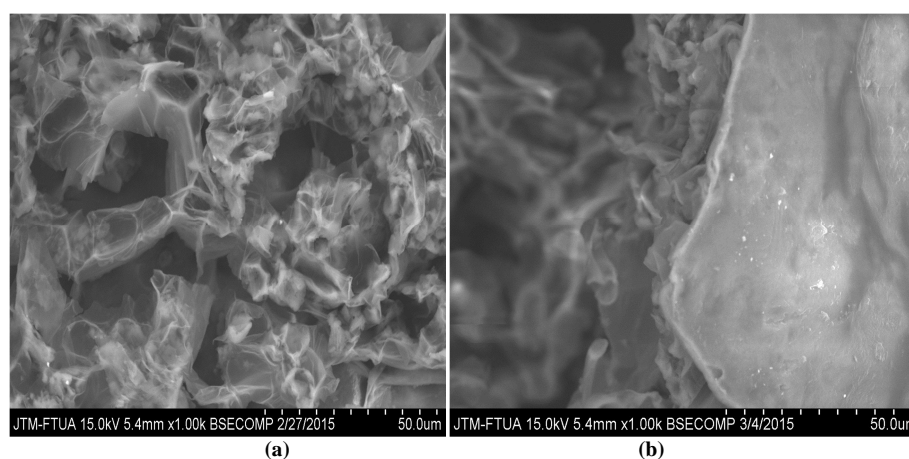


Figure 6. SEM image of *Annona muricata* L leaves at 1000 x magnification: (a) before uptake of Cu (II); (b) after uptake of Cu (II)

After the biosorption process, the SEM image of *Annona muricata* L leaves revealed the combination of small and particles size which was suggested that to be an appropriate structure for metal ion concentration.

## CONCLUSION

The study indicated that *Annona muricata* L leaves can be used as an effective adsorbent material for Cu (II) in aqueous solution. Biosorption of Cu (II) by using soursop *Annona muricata* L leaves is known to be dependent with pH, initial concentration of Cu (II) ions, biosorbent dosage, and contact time. The optimum pH for biosorption process was pH 7, with initial concentration 500 mg/L, biosorbent dosage 0.25 g and contact time 90 min. From FTIR analysis it is estimated that the functional groups that involved in biosorption process are O-H and N-H groups. The SEM image shows that *Annona muricata* L leaves have a porous surface, indicated that it have good potential as biosorbent.

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