Biosorption of Copper from Aqueous Solution by Algal Biomass

Z. Al-Qodah, Eman Assirey

Abstract

Sorption experiments have been done using the dead biomass of Spirogyra species, copper removal are carried out to study the influence and optimization of the biosorption. The effects of pH, initial metal ion concentration, different contact periods, and varying biomass quantities have been also studied. It is observed that Spirogyra species possess good specific uptake capacities for Cu (II). Spirogyra species has shown much better sorption in the pH between 6.0 and 7.0, and at contact time of 80 min. The equilibrium isotherm has been analyzed using Freundlich and Langmuir isotherm models and is best described by the Langmuir model.

Keywords: [copper removal, Spirogyra, Adsorption isotherms]

1. Introduction

The remarkable increase of industrial processes in addition to many human activities that use heavy metals has intensified environmental contamination and pollution problems [1]. As a result of their mobility and solubility, the accumulation of heavy metals in the environment leads to a major problem face by the society from one hand [2, 3] and to the deterioration of many ecosystems from another hand [4]. As a consequence, there are ever increasing legislative standards in most countries that impose treatment processes to reduce heavy metals concentration or to recover them where feasible. The metals ions of primary concern according to World Health Organization (WHO) are those of aluminum, cadmium, chromium cobalt, copper, iron, manganese, mercury, lead and nickel, [4, 5].

Copper, Cu is considered as one of the most toxic and widely used material, since it is involved in variety of industries including such as electronic and electrical, metal plating, mining, manufacture of computer, a component in ceramic glazing and glass coloring. Those industries and others discharge a huge amount of wastewater contaminated with a significant amount of copper [2-3, 6-7]. Notably, Cu^{+2} ions are considered as persistent, non-degradable, bio-accumulative and toxic chemical species. In spite that Cu^{+2} is essential to both human and plant life and health as well. This ion has many adverse effects on the environment and human health. In humans, Cu^{+2} concentration above 0.05 mg/l, can cause serious medical problems including severe mucosal irritation, anemia, stomach intestinal distress, central nervous problems followed by depression and kidney damage with prolonged exposure [8-13]. Accordingly, the strict environmental regulations have enhanced the search for new and efficient and environmentally friendly processes for metal ions removal from wastewater to attain toxicity the maximum allowable limits [1, 3, and 14].

For this reason, many processes for Cu^{+2} ion removal have been applied in the last two decades. These include evaporative recovery, ion exchange, reverse osmosis, electrochemical treatment and adsorption. However, the application of most of these processes is often limited due to technical or economic constraints [15]. Among these processes, adsorption is highly recommended because it is proven as a simple, economical, effective and environmental friendly process [16]. Among various adsorbents used, activated carbon is considered as the most efficient material used due several important properties that enhances the adsorption process. These properties include the high surface area, friendly behavior to the environment and ease in operation [17]. However, activated carbon is economically ineffective. For this reason, this drawback has led to the search for suitable cheap and efficient adsorbents [18].

Recently, several types of bio-adsorbents including: some agricultural wastes, living and non-living fungi, algae and bacteria have been used as efficient and low cost alternatives [18-22]. However, the use of non-living cells as metal binding bio-adsorbents has been gaining advantages and becomes more attractive and practical then living cells. This is referred to the fact that living cells will be deactivated by toxic heavy metals ions, resulting in cell death [23]. Moreover, living cells usually grow in a fermentation medium that contains nutrients. These nutrients increase the BOD and COD in the effluent [24]. Moreover, both the adsorbed metals ions and the biomass can be easily recovered and regenerated and reused using suitable chemical and physical processes, leading to repeated use of the biomass and better process economy [25].

Algae, is a cheap and available filamentous microorganism obtained from marine or freshwater has been successfully used as a bio-adsorbent for heavy metal ions from industrial wastewater [25-27]. However, very limited research has been conducted to utilize algae as a bio-adsorbent for copper ion, Cu^{+2}. Bishnoi et al., [5] used dead iomass *Spirogyara* algal species for the adsorption of Cu^{+2}. The bioadsorption result were fitted to Freundlich isotherm model and the maximum adsorption capacity reached about 35 mg Cu^{+2} / g adsorbent. Subsequently, Abu Al-Rub et al., [3] used a powder of *Chlorella vulgaris* algal species for copper Biosorption. The experimental results were fitted to Langmuir, Freundlich and Radushkevich isotherm models at different values of pH. They reported that maximum adsorption capacity was dependent on pH with a maximum value of 55 mg /g adsorbent at pH =5. Vilar et al., [28] used two an algal waste from the agar extraction industry, either in its original form or granulated by polyacrylonitrile and the marine red algae *Gelidium* for the bioadsorption of Cu^{+2}. They found that a non-ideal, semi-empirical, thermodynamically consistent (NICCA) isotherm fitted their experimental results with r algae *Gelidium* for different pH values and copper concentrations. Recently, Ahmady-Asbchin and Mohammadi [29] studied the bioadsorption properties of Cu^{+2} by a pre-treated biomass of marine algae *Fucus vesiculosus*. They reported that the adsorption equilibrium data were best fitted by the Langmuir's isotherm model. Li et al., [30] studied the bioadsorption of Cu^{+2} found in brewery waste by red *Palmaria palmate* algae. They found that the process was was strongly dependent on the pH value and initial Cu^{+2} concentration. In addition, Langmuir isotherm model described well the experimental results better than other models. In a more recent study, Al-Homaidan et al., studied Cu^{+2} from aqueous solutions bioadsorption by the micro-alga (cyanobacterium) *Spirulina* *platensis*. They reported that the maximum Cu^{+2} uptake by the micro algae was observed at pH 7 where a maximum of 90.6% ion removal from a solution containing 100 mg Cu^{+2} / L, at pH 7, with 0.050 g dry biomass and at 37 ^{o}C with 90 min of contact time..

It is evident from the above survey that all types of algae represent a potential adsorbent for heavy metals and especially copper ions. However, there is a need for more research to investigate the kinetic and isotherm models for this process and to link the obtained results with the algae micro structure in addition to the operational parameters. For this reason, the objective of this investigation is to use dead algae cells as a bio-adsorbent for copper ion, Cu^{+2}. Both kinetic and isotherm models will be developed to fit the experimental data. The effect of different operational parameters such as temperature, pH, bio-adsorbent mass and Cu^{+2} initial concentrations will be investigated.

Biosorption, is considered as one of the most promising technologies involved in the removal of toxic metals from industrial waste streams and natural waters (5,6). Biosorption is a term that describes the removal of Heavy metals by the passive binding to non-living biomass from an aqueous solution. There are lots of studies showed that algae, bacteria, fungi or higher plants that uptake or accumulate large amounts of heavy metals from their environment (7-9).

Biosorption is possible by both living and non-living biomass; there are two modes of metal ion uptake. The first mode is independent of cell metabolic activity, and is referred to as biosorption or ions to cell wall. This mode is common to both living and dead cells. The second mode of metal uptake into the cell across the cell membrane is dependent on the cell metabolism, and is referred to as intracellular uptake, active uptake or bioaccumulation. This mode occurs in living cells only. In this study biosorption of copper ion from aqueous solution was investigated using green Spirogyra algae as an adsorbent. A series of experiments were conducted at different operational conditions (9).

Recently, bioadsorbents have emerged as an eco-friendly, effective and low cost material option [5,9,11]. These bioadsorbents include some agricultural wastes, fungi, algae and bacteria [12,13].

Studies using bioadsorbents have shown that both living and dead microbial cells are able to adsorb metal ions and offer potential inexpensive alternative to conventional adsorbents [14,15]. However, living cells are subject to toxic effect of the heavy metals, resulting in cell death [16]. Moreover, living cells often require the addition of nutrients and hence increase the BOD and COD in the effluent [17]. For these reasons, the use of nonliving biomaterials or dead cells as metal binding compounds has been gaining advantage because toxic ions do not affect them. In addition, dead cells require less care, maintenance and they are cheaper [18]. Furthermore, dead biomass can be easily regenerated and reused [19].

1.1 Isotherm models

Sorption isotherms indicate molecules distribution between the liquid and the solid phase when the sorption process reaches equilibrium state. To further explore the adsorption mechanism, Langmuir and Freundlich isotherm models were used to analyze the adsorption data. The analyses of the isotherm have been established by fitting data to both isotherm models to find the suitable model. The linearized Langmuir isotherm is represented by the following linear equation (10):

$$1/Q_e = 1/Q_{max} + (1/bC_e+1)$$

Where Ce (mg/L) is the equilibrium concentration, Q_e (mg/g) the amount of adsorbate adsorbed or desorbed per unit mass of adsorbent, and Q_m and b are the Langmuir constants related to sorption capacity and rate of sorption, respectively. The essential characteristics of Langmuir can be expressed by a dimensionless constant called separation factor or equilibrium parameter R_L, defined as (11)

$$R_L = 1/(1 + (bC_o))$$

Where b is the Langmuir constant and C_0 (mg/L) is the initial ion concentration. The value of R_L give indication about the type of the isotherm to be either unfavorable (R_L >1), linear (R_L = 1), favorable ($0<R_L<1$) or reversible (R_L = 0).

The linear form of the Freundlich equation is:

$$\mathrm{lnQe} = lnKf + \frac{1}{n} + \mathrm{lnCe}$$

Where Qe is the amount adsorbed or desorbed at equilibrium (mg/g) and Ce (mg/L) is the equilibrium concentration. KF and n are Freundlich constants, n gives an indication of how favorable the sorption process and K_F is the sorption capacity of the adsorbent. The slope 1/n ranging between 0 and 1 is a measure of sorption intensity or surface heterogeneity. The surface become more heterogeneous as its value gets closer to 0 (11).

2. Materials and Methods

2.1 Preparation of biomass.

Algal biomass was collected from a pool to irrigate crops in (Alrsaifah-Jordan), where these are available in abundance. After sample collection, it was thoroughly washed with tap water to remove dirt and other unwanted material, and then washed with distilled water, and sample was squeezed, then water decanted and sample was dried at 90 °C for six hours in a drying oven, after that biomass sample has been grinded using mortar. Then grinded sample has been sieved and particles with 300 μm diameter have been selected.

2.2 **Preparation of copper solution.**

Cu stock solution has been prepared from $CuSO_4.5H_2O$; pH has been adjusted using HCl 1.0M and NaOH 1.0M

2.3 **Biosorption process.**

0.50g —2.00g of biomass powder was contacted with a 100.0 mL several known concentrations of copper solution (50-300 ppm) with stirring. Samples were taken out at specific duration of time and centrifuged at 5000 rpm for 10 min using (Hettich) centrifuge device.

2.4 Analysis of Cu (II) ions

Determinations of Cu (II) ions concentration have been done using spectrophotometer (JENWAY PCO1) at wavelength 580 nm.

3. Results and Discussions

3.1 Analysis of Cu (II) ion

Figure 1 shows standard calibration curve of Cu (II), spectrophotometric analysis has been done at wavelength 580 nm, high R^2 reflects high correlation in the study targeted Cu (II) concentration range.

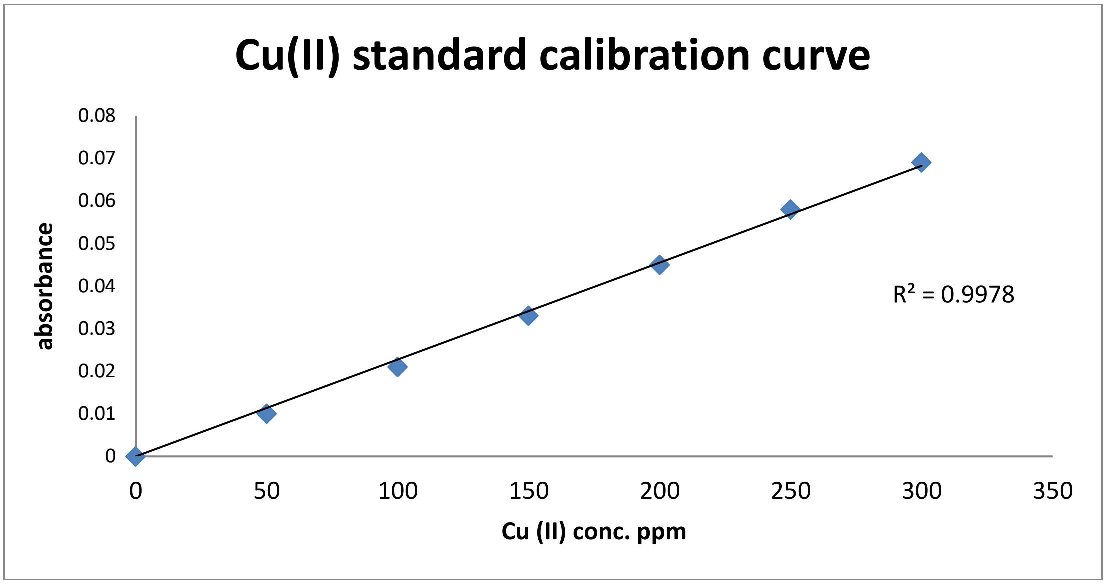

Fig. 1: Standard calibration curve of Cu (II) at 580 nm

3.2 Results of Biosorption Process
3*.2.1 Copper – biomass contact time study:

The contact time has been studied for 0.5 and 1.0g algal dose with several Cu (II) initial concentrations (50-300ppm), adsorption of Cu (II) reach equilibrium after 80 minutes as shown in figure 2, almost other initial concentration have reached the equilibrium at the same time.

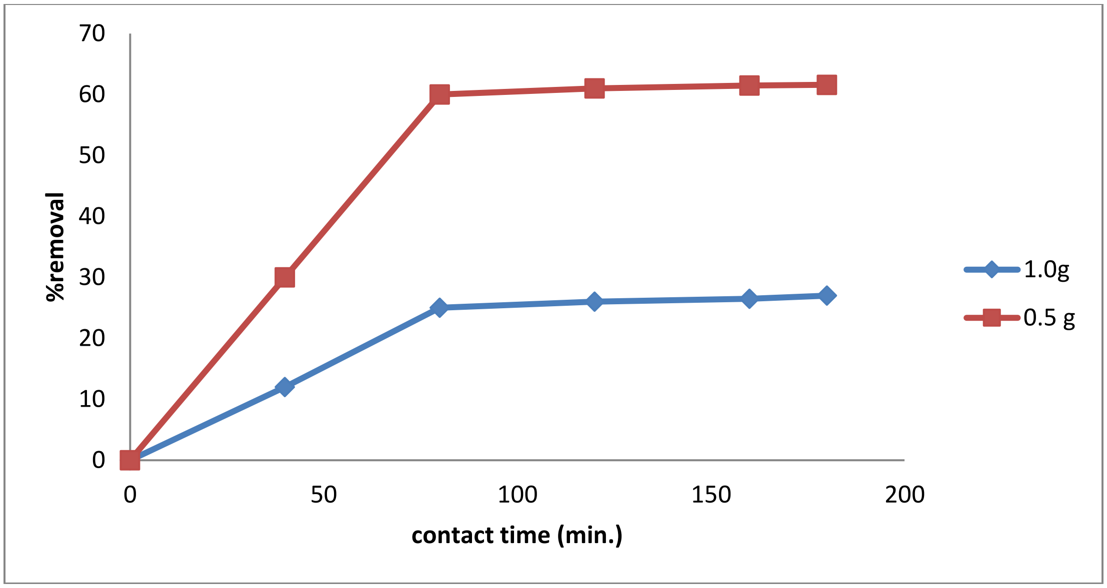

Figure 2: Effect of contact time on percent removal Cu (II) by 0.5 and 1.0g green algae, at initial Concentration = 50 ppm and pH 7

3.2.2 Effect of varying biomass dose:

Effect of adsorbent dose has been studied by varying the amount of adsorbents from 0.5-1.0 g at pH 7.0. Results shown in Figure 2 reflect that the maximum removal of Cu (II) is obtained at minimum dose of 0.5g, i.e with increase in dose; there was decrease in percentage removal from (60%-25%). Copper uptake decreases when biosorbents *concentration increases. The higher biomass causes screen effect of dense outer layer of cells, blocking the binding sites from metal ions, resulting in lower metal removal per unit biomass.

3.3 Effect of initial metal ion concentration:

Different initial Cu (II) ion concentrations at adsorbent dose of 0.5 g, contact time of 80 minutes, and pH 7.0 have been studied. As shown in Figure 3. It is clear from Figure 3 that as the concentration of Cu (II) increases the percentage removal efficiency increase.

This increase was observed from (60%- 80 %) during concentration, the highest removal at 150 ppm, then the percent removal decreases with increase in concentration, because of the reaching to the threshold of adsorption capacity.

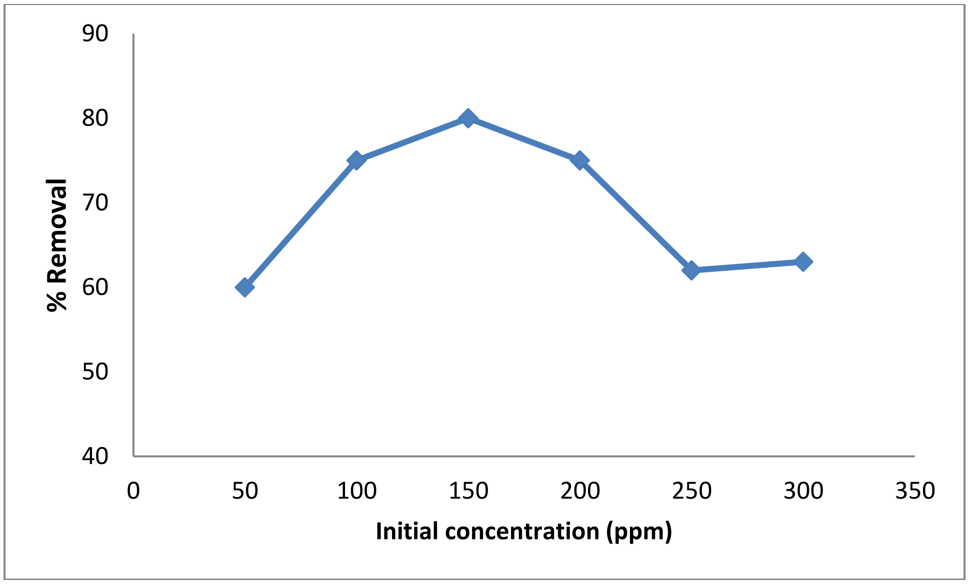

Figure 3: effect of initial Cu (II) concentration on its percent removal by green algae

3.4 Effect of pH on adsorption:

To evaluate the effect of pH on the adsorption behavior of Cu (II) on algal, Cu (II) solutions have been adjusted to several pH values as shown in Figure 4. An increase in percent removal with increase in pH of the medium was observed for the metal ion up to pH value of 7.0. Then there was decrease in percent removal above this pH. It has been indicated that on increasing the pH from (1-7) of the medium the sorption capacity increased from (31%-86%).

The results suggest that the adsorption of Cu to the biomass is mainly due to ionic attraction. Therefore, as pH decreases the cell surface becomes more positively charged, reducing the attraction between biomass and metal ions. In contrast, higher pH results in facilitation of metal uptake, since the cell surface is more negatively charged.

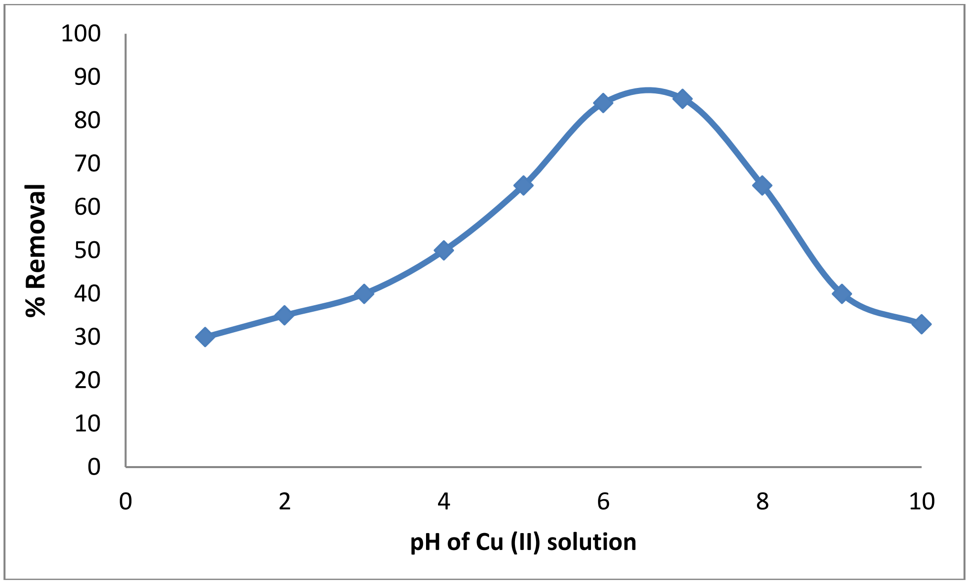

Figure 4: effect of pH on percent removal of Cu (II) by green algae

3.5 Sorption isotherms:

Figures 5 and 6 illustrates that the adsorption of Cu^{2+} ions fitted well with both the Langmuir and the Freundlich isotherm models, but based on the values of the statistical parameters for both models, it can be concluded that Langmuir models provide better representation of the experimental data, followed by Freundlich model. The Langmuir and Freundlich isotherm model constants are given in Table 1.

The Langmuir isotherm assumes the mechanism of the adsorption process as a monolayer adsorption on the surface of the adsorbent:

. Figure 5 illustrates the Langmuir plot of 1/qe versus 1/Ce, enabling the calculation of Langmuir constants from the intercept and slope of the linearized plot. Freundlich isotherm model, on the other hand, describes the adsorption occurring on heterogeneous surface with uniform energy; kf and n represent the Freundlich constants, which can be calculated from the slope and intercept of the linear plot of ln qe versus ln Ce as presented in Figure 6.

For the Langmuir isotherm model, separation factor or equilibrium parameter (R_L) can be used to describe the favorability of adsorption on the polymer surface Table 2.

The R_L values for the adsorption of copper metal ions are given in Table 2, which reveals

that the values fall in the preferred region (i.e., $0 < RL < 1$). The results thus certify that algal biomass is a good adsorbent for the removal of heavy metal ions in aqueous solutions.

Table 1. Langmuir and Freundlich Isotherm Model Constants for Cu^{2+}Adsorption

Langmuir isotherm model		
Q_m	b	R^2
49.50	0.240	0.976
Freundlich Isotherm Model		
K_f	n	R^2
2.606	1.716	0.894

Table 2. Separation factor (Langmuir isotherm)

C_o	R_L
50	0.453
100	0.293
150	0.216
200	0.171
250	0.142
300	0.121

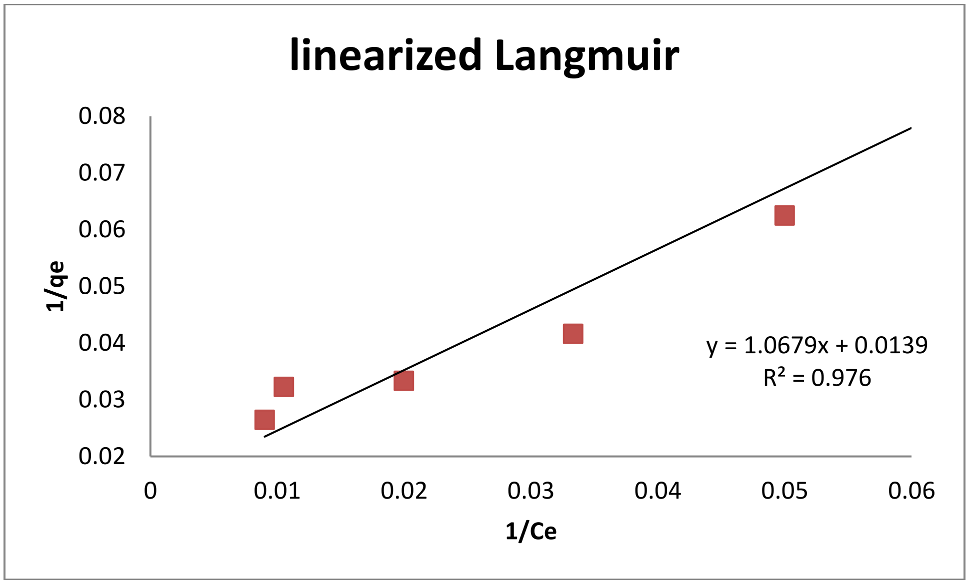

Figure 5:Langmuir isotherm of the adsorption of Cu^{2+} ions on algal biomass

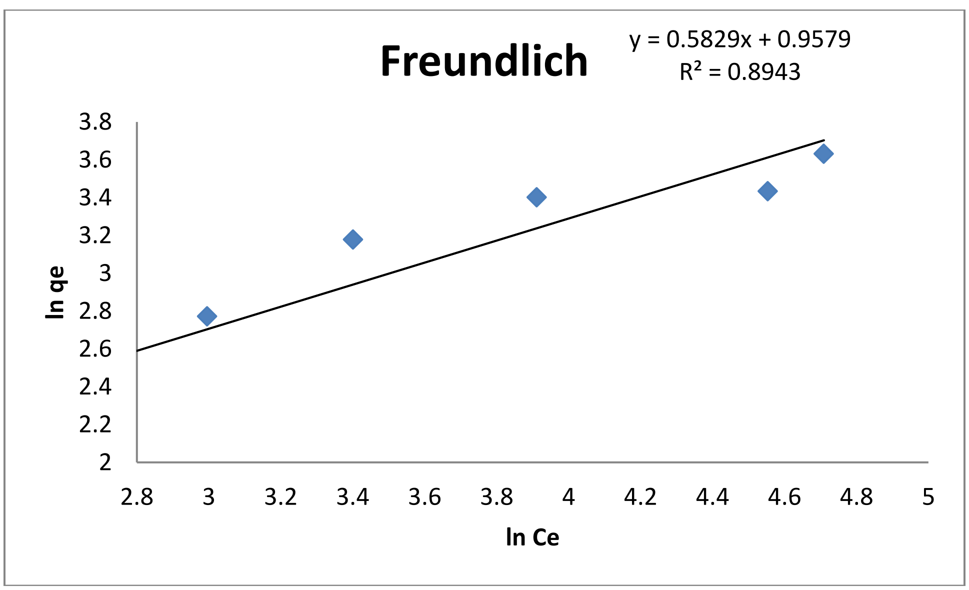

Figure 6: Freundlich isotherm of the adsorption of Cu^{2+} ions on algal biomass

4. Conclusion:

In this paper biosorption of copper ion from aqueous solution was investigated using green algae as an adsorbent. A series of experiments were conducted at different operational conditions. Different parameters affect the copper ion adsorption process; the initial rate of adsorption was greater for higher ion initial concentration. As the adsorbent mass dose increase the ratio of the final concentration to the initial concentration decrease; so the higher adsorbent mass has a higher ability to remove the ions from the aqueous solution. pH around 6 natural pH showed the highest adsorption capacity. Langmuir model provide the best representation of the experimental data, followed by the Freundlich model.

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