Journal of Hazardous Materials 173 (2010) 502–509

Contents lists available at ScienceDirect

Journal of Hazardous Materials

journal homepage: www.elsevier.com/locate/jhazmat

Removal of Pb(II) ions from aqueous solution by adsorption using bael leaves (*Aegle marmelos*)

S. Chakravarty*, Ashok Mohanty, T. Nag Sudha, A.K. Upadhyay, J. Konar, J.K. Sircar, A. Madhukar, K.K. Gupta

Analytical Chemistry Centre, National Metallurgical Laboratory, Jamshedpur 831007, Jharkhand, India

ARTICLE INFO

Article history:
Received 5 May 2009
Received in revised form 19 August 2009
Accepted 24 August 2009
Available online 31 August 2009

Keywords:
Biosorption
Aegle marmelos
Pb(II) ions
Aqueous solution

ABSTRACT

Biosorption of Pb(II) on bael leaves (*Aegle marmelos*) was investigated for the removal of Pb(II) from aqueous solution using different doses of adsorbent, initial pH, and contact time. The maximum Pb loading capacity of the bael leaves was 104 mg g^{-1} at 50 mg L^{-1} initial Pb(II) concentration at pH 5.1. SEM and FT-IR studies indicated that the adsorption of Pb(II) occurs inside the wall of the hollow tubes present in the bael leaves and carboxylic acid, thioester and sulphonamide groups are involved in the process. The sorption process was best described by pseudo second order kinetics. Among Freundlich and Langmuir isotherms, the latter had a better fit with the experimental data. The activation energy E_a confirmed that the nature of adsorption was physisorption. Bael leaves can selectively remove Pb(II) in the presence of other metal ions. This was demonstrated by removing Pb from the effluent of exhausted batteries.

1. Introduction

Industrial effluents are loaded with heavy metals which are hazardous to humans and other forms of life. Their toxic nature has a severe environmental impact. This has resulted in the enforcement of stringent laws for the maximum allowable limits of their discharge into the open landscapes and water bodies. Lead is used as an industrial raw material for storage battery manufacturing, printing, pigments, fuels, photography material and explosive manufacturing [1]. Since Pb(II) is a potent neurotoxic metal, its pollution is of major concern. The presence of lead in drinking water, even in low concentrations, may cause diseases such as anemia, hepatitis, nephrite syndrome, etc [2].

A number of processes exist for the removal of metal pollutants from waste water, viz. precipitation, electroplating, ion exchange and membrane separation. These processes have several disadvantages such as incomplete metal removal, high reagent and energy requirement and generation of toxic sludge/waste products that require disposal and further treatment. This led to the need to develop an effective, low cost and environment friendly process for wastewater treatment. Adsorption is an efficient and economical process used for the removal of heavy metals from industrial wastewater. A number of investigations have been carried out using naturally occurring waste materials for the removal of lead from aqueous systems. Some of the naturally occurring waste materials used for this purpose are fly ash [3], phosphatic clay [4], clonoptilolite [5] and sea nodule [6]. Many biological materials such as olive stones, fungus *Aspergillus niger*, seaweeds have also shown potential for the removal of heavy metal [7]. Biosorption of Pb(II) onto a cone biomass of *Pinus sylvestris* was studied which revealed that the maximum adsorption of Pb onto *P. sylvestris* was at pH 4.0 [8]. In another study, ten different seaweed species were compared on the basis of lead uptake at different pH conditions [7]. The study revealed that the brown seaweed, *Turbinarea concoides* exhibited the maximum lead uptake of 439.4 mg/g at an optimum pH of 4.5 and temperature of 30 °C. Brown seaweed was extensively used for biosorption and its polysaccharide content was believed to be responsible for its excellent metal binding capacity [9]. It was observed that the biological materials could accumulate heavy metals in their walls, even though the binding sites for chelating were not identified [10].

The objective of the present study is to investigate the utility of various types of locally available biomass to remove Pb(II) from aqueous systems. Dry leaves of bael tree (*Aegle marmelos*) were tested for the removal of Pb(II) ion from aqueous medium as they are very cheap and easily available biomass. Bael tree is a spiny tree belonging to the family Rutaceae and is abundantly found in India, Myanmar, Pakistan and Bangladesh. Literature reports indicate that the leaves of bael tree have medicinal value and are useful for the treatment of ophthalmia, deafness, inflammations, cataract, diabetes, diarrhoea, dysentery, heart palpitation and asthmatic complications [11]. In the present study, leaves of the bael

* Corresponding author. Tel.: +91 657 2270588; fax: +91 657 2345213.
E-mail address: sanchita@nmlindia.org (S. Chakravarty).

0304-3894/$ – see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.jhazmat.2009.08.113

tree were successfully utilized for the removal of Pb(II) ions from aqueous solution. The loading capacity of the bael leaves was optimized and the selectivity of the bael leaves towards Pb(II) in the presence of other metal cations was investigated. Attempts have been made to understand the factors responsible for adsorption of Pb(II) to the bael leaves. In an effort to show its practical utility, bael leaves were utilized for the removal of Pb(II) from storage battery effluents at the laboratory scale. This paper aims to examine the use of bael leaves as an adsorbent for the contaminated water bodies.

2. Experimental

2.1. Biosorbent

In the present study, bael leaves of the Indian bael tree (*Aegle marmelos*) were used as a biosorbent for the removal of Pb from aqueous solution. The bael tree is native to northern India but is found widely throughout the Indian peninsula and in Ceylon, Burma, Thailand, Indo-China [12]. The bael leaves were first washed thoroughly with deionised water. The soluble colored components were then removed by washing repeatedly with hot deionised water (70 °C). The bael leaves were then dried at 70 °C for 24 h. The dried bael leaves were crushed and sieved through 100 mesh and stored in polythene bottles.

2.2. Instrumentation

Atomic absorption spectrometer (AAS), GBC, AVANTA equipped with an air acetylene burner was used to determine the concentration of all the metal ions in the aqueous solution. The hollow cathode lamp was operated at an analytical wavelength of 283.3 nm for Pb and the slit was 0.2 nm for all the experiments. FT-IR spectra of bael leaves samples (before and after adsorption) were obtained using FT-IR spectrometer, Thermo Nicolet, Nexus 870. The processed dry bael leaves in powder form (Section 2.1, about 0.1 g) along with KBr were ground into fine particles and pressed to make pellets. FT-IR spectrum of the native bael leaves was then recorded using the pellets. For FT-IR spectrum of Pb loaded bael leaves, the native bael leaves were adsorbed with 1000 mg L^{-1} Pb(II) solution for 24 h. After adsorption, Pb(II) loaded bael leaves were filtered, washed with deionised water and air dried. The FT-IR spectrum of Pb loaded bael leaves was then recorded using KBr pellets. A Scanning Electron Microscope, JEOL JSM 840, was used to obtain SEM images of the bael leaves before and after Pb adsorption. Metal mapping of the native bael leaves and Pb loaded bael leaves were performed using the same SEM. All pH measurements were made using a digital pH meter.

2.3. Metal solutions

All the chemicals used were AR grade supplied by SRL, Mumbai, India and deionised water was used for preparation of the solution throughout the experiments. The stock solution of Pb(II) was prepared by dissolving $Pb(NO_3)_2$ in deionised water. Stock solutions of Mn^{2+}, Fe^{2+}, Co^{2+}, Ni^{2+}, Cu^{2+}, Ca^{2+} and Mg^{2+} were prepared by dissolving their corresponding nitrate salts in deionised water. The working solutions were prepared by appropriate dilution of the stock solutions with deionised water.

2.4. Batch adsorption studies

Batch adsorption studies were performed in 100 mL conical flasks to optimize the operating conditions for Pb(II) adsorption by bael leaves. In a typical experiment, 0.2 g of processed powder bael leaves (Section 2.1) were added to 50 mL of 98 mg L^{-1} synthetic Pb(II) solution. The natural pH of the synthetic Pb(II) solutions was measured to be 5.1. This pH was maintained throughout for all the experiments except for pH optimization studies. The mixture was shaken for 45 min at 303 K using a mechanical shaker at 120 rpm. The phases were separated by filtration and the Pb(II) concentration in the filtrate was determined by Atomic Absorption Spectrophotometer (AAS). Adsorption parameters, viz. biosorbent dose, initial concentration of Pb(II) and contact time were optimized by continuous variation method. For pH optimization, the initial pH of each Pb(II) solution was adjusted to the required pH by using 0.01 N HNO_3 or 0.01 M NaOH. The concentration of Pb was measured using AAS after the pH adjustment. For desorption studies, Pb(II) was initially adsorbed on the bael leaves at pH 5.1. The equilibrium concentration (C_e) of Pb in the solution was measured and the pH was then adjusted to the range of 2.0–7.0 using 0.01 M NaOH or 0.01 N HNO_3. The solution after pH adjustment was shaken for half an hour and the equilibrium concentration after desorption (C_e') was measured. The percentage of desorption was calculated as follows:

$$\%\text{desorption} = \left(\frac{C_e' - C_e}{C_o - C_e}\right) \times 100$$

where C_o is the initial Pb(II) concentration of the solution (mg L^{-1}), C_e is the equilibrium Pb(II) concentration (mg L^{-1}) at a particular pH and C_e' is the equilibrium Pb(II) concentration (mg L^{-1}) at a particular pH after desorption. The kinetic studies were carried out at three different Pb(II) concentrations of 48.2, 87.8 and 180.2 mg L^{-1} at temperatures of 303, 313 and 323 K.

3. Results and discussion

3.1. Removal of Pb(II) from synthetic solutions

Dry bael leaves powder was initially used to remove Pb(II) from synthetic aqueous solutions. Different experimental parameters like contact time, adsorbent dose, and pH were optimized to obtain maximum Pb(II) removal using the bael leaves. For optimization of contact time, a solution of 98 mg L^{-1} Pb(II) at pH 5.1 with 0.2 g of bael leaves was used. The contact time was varied between 5 and 120 min at three different temperatures of 303, 313, and 323 K (results not shown here). It was observed that during the first 20 min there was a rapid uptake of Pb, up to a maximum of 65%. The time required for attaining equilibrium was 30 min. Therefore, a contact time of 30 min was employed for all other studies. It is to be noted that a contact time of 30 min was sufficient for optimum removal of Pb(II) when the initial Pb(II) concentration was raised to 180.2 g L^{-1}.

3.1.1. Effect of adsorbent dose

Three different solutions having an initial Pb(II) concentration of 48.2, 87.8 and 180.2 mg L^{-1}, with pH 5.1 were used to study the effect of the adsorbent dose on the removal of lead. The adsorbent dose was varied in the range of 0.01–0.5 g. The result is presented in Fig. 1. It was observed that the removal efficiency increased with increase of adsorbent dose. It reached a maximum (85% approximately) at around 0.2 g and then remained almost constant. Therefore, the optimum bael leaves dose was taken as 0.2 g and this was used for all further studies. The positive correlation between adsorbent dose and metal removal efficiency can be related to the increasing surface area of the available binding sites [13].

3.1.2. Effect of pH

pH is a very important parameter that affects any biosorption process. It affects the activity of the functional groups present in the biosorbent that are responsible for metal adsorption and also affects the competition of metallic ions to get adsorbed to the