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Development of hybrid membrane process for Pb bearing wastewater treatment

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ABSTRACT

In present study, two hybridization scheme, (i) biosorption process followed by NF/RO membrane process and (ii) NF/RO membrane process followed by biosorption process were studied with the aim of developing a hybrid separation process for the efficient and cost effective removal of Pb ions from aqueous solution. Two polyethersulfone (PES) NF and RO membranes were used in a rectangular cross flow membrane cell. The influence of membrane process parameter such as trans-membrane pressure and cross flow velocity on permeate flux and Pb ion rejection were investigated. In biosorption process, a low cost novel biosorbent *Trapa rossica* leaves powder was used and tested for their Pb ion binding capacity. A set of batch biosorption experiments were conducted to evaluate the effect of biosorption process parameters on Pb ion uptake. The biosorption results indicated the potential of *T. rossica* leaves as biosorbent and can be used efficiently for the treatment of Pb ion bearing wastewater at lower concentration. From hybrid processes, it was found that biosorption process followed by NF membrane process was most preferable because it gave higher permeate flux as well as higher Pb ion rejection.

1. Introduction

The presence of heavy metal ions in aqueous systems is one of the serious worldwide environmental problem. Heavy metal ion contaminated water cause adverse effect on ecosystem particularly human health because of their toxic nature. Heavy metals, such as Arsenic, Beryllium, Cadmium, Chromium, Copper, Lead, Lithium, Mercury, Nickel, Tin and Zinc are among the most toxic heavy metal ions that are used in various industries, such as automotive, ceramic, coating, electroplating, mining, refineries, storage batteries, smelters and textile industry. Among all, Pb ion is highly toxic and affects human health even at very low concentration exposure. The maximum exposure limit of Pb ion is 0.05 mg L^{-1} set by WHO [1]. Prolonged exposure to Pb ions causes damage in brain, liver, kidney, nervous system and reproductive system of children and pregnant women [2,3].

Available conventional treatment methods used for treatment of heavy metal ion contaminated wastewater includes, coagulation and flocculation, chemical precipitation, adsorption, chemical oxidation, ion exchange and electrochemical treatment. These processes have significant disadvantages such as huge amount of sludge formation which creates secondary pollution, incomplete removal of metal ion, time consuming and requirement of expensive equipments and chemicals and consumes high energy [4–6]. Biosorption process is another potential technique of wastewater treatment, which was frequently studied by many researchers because of its advantage such as low cost and environment friendly [7–10]. Biosorption process utilizes inactive or dead biomass of living or nonliving organism as adsorbent which are capable of binding the heavy metal ions on its surface [11–13]. Similar to biosorption, membrane processes are highly efficient and widely used technique in industries to remove heavy metal ions as well as other pollutants from wastewater [14,15]. Hajdu et al. have reported 90–100% removal of Pb ion with poly-gamma-glutamic acid (γ-PGA) nanoparticle enhanced ultrafiltration technique using a membrane made of regenerated cellulose having molucular weight cut-off of 10 kDa and membrane area of 50 cm^2 used with the Pellicon XL device [16]. However, membrane fouling due to blocking of membrane pores by metal ions, lowers the flux and demands increased energy requirement, which lead to high operating cost, is the major concern of membrane processes. In order to reduce membrane fouling, increase the membrane flux and to increase the removal of pollutant from wastewater; in many studies membrane process was coupled with conventional wastewater treatment process [17–21]. Hybrid membrane processes have shown advantages over conventional processes such as higher removal efficiency, high quantity of treated wastewater, better fouling control, low energy consumption and lower backwashing time [22].

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Nomenclature

A	Membrane area in m^2
J	Calculated permeate flux in $m^3 m^{-2} s^{-1}$
t	Time in sec
v	Permeate volume in m^3
$1/n$	Freundlich model constants related to intensity of adsorption
B	Constant related to adsorption energy E in $mol^2\ kJ^{-2}$
E	Free energy of adsorption in $kJ\ mol^{-1}$
e	Polanyi potential
q	Metal uptake capacity in $mg\ g^{-1}$
R	Gas constant in $kJ\ mol^{-1}\ K^{-1}$
T	Temperature in K
V	Volume of the solution in L
W	Mass of biosorbent in g
C_e	Equilibrium metal ion concentration in $mg\ L^{-1}$
C_i	Initial metal ion concentration in $mg\ L^{-1}$
C_o	Initial metal ion concentration in $mg\ L^{-1}$
K'	Pseudo-second-order rate constant of biosorption in $mg\ g^{-1}\ min^{-1}$
K_d	First-order lagergren kinetics rate constant of biosorption in min^{-1}
K_F	Freundlich model constants related to adsorption capacity
K_L	Langmuir biosorption constant in $L\ mg^{-1}$ relating the free energy of biosorption
K_p	Intra-particle diffusion rate constant in $mg\ g^{-1}\ min^{-0.5}$
q_e	Equilibrium metal uptake capacity in $mg\ g^{-1}$
q_m	Monolayer biosorption capacity of the biosorbent in $mg\ g^{-1}$
q_t	Metal uptake capacity in $mg\ g^{-1}$
R_L	Dimensionless constant separation factor

In the present work, an alternative process for the cost efficient removal of Pb ions by coupling of biosorption process with NF/RO membranes in various schemes was investigated. First scheme was biosorption followed by NF/RO process and the second scheme was NF/RO process followed by biosorption. For the biosorption process, leaves of *Trapa rossica* also known as pani singhara, pani phal, water caltrop, water chestnut and buffalo nut was used as biosorbent in this study. *T. rossica* is annual aquatic plant grows in slow moving or stationary water and have been cultivated in India for edible seeds. After taking the fruits, the plant remains as waste in water system. Biosorption kinetics and mechanism for this adsorbent have been discussed and also a comparison of biosorption process, membrane filtration process and hybrid process have been reported.

2. Materials and methods

2.1. Reagents

Lead nitrate ($Pb(NO_3)_2$) purity >99% (Merck, Germany), HNO_3 ACS, R (Merck, Germany), NaOH (Merck, Germany), potassium bromide (KBr) (Merck KGaA, Germany). The chemicals and reagents used in experiments were of analytical grade and used without any further purification.

2.2. Feed (synthetic wastewater)

1000 mL stock solution of Pb ion contaminated wastewater was prepared by dissolving 1.599 g purified lead nitrate in approximately 200 mL of deionized water. 10 mL of 1.0 M HNO_3 was added and was shaked for 15 min for complete dissolution and diluted to 1000 mL with deionized water. The stock solution was stored in a volumetric flask and used after further dilution ranging between 10 and 200 $mg\ L^{-1}$ concentrations as required in experiments. Deionized water was used as dispersant for all the experiments.

Table 1
The physical characteristics of the *T. rossica*.

Parameter	Value
Bulk density	0.38 $g\ mL^{-1}$
Porosity	46%
BET surface area	11.45 $m^2\ g^{-1}$
Average particle size	106 μm
Moisture content	78%
Point of zero charge	5.4

2.3. Biosorbent

The adsorbent *T. rossica* leaves were collected from a pond outside IIT Guwahati campus and washed with tap water to remove adhering dust particle and soluble impurities from the leaves surface. After drying in sun light for 6 days it was dried in hot air oven at 60 °C for 36 h. The dried leaves were grinded using mixer grinder (Bajaj mixer grinder, India) to make it fine powder. This biosorbent was preserved in zip lock bags after sieving through 140 ASTM E-11 sieves (particle size 106 μm).

2.4. Membrane

Two types of membranes were flat sheet membranes used in this study, one thin film composite NF membrane of MWCO 400 Da and another is a RO membrane. Both the membranes were made from polyethersulfone (PES) and were purchased from M/s Permionics Membranes Pvt., Ltd., Gujarat, India. The effective surface area of cross flow membrane module was 0.0164 m^2. Permeate water were collected at different times interval for residual Pb ion analysis and values of permeate flux was determined by using Eq. (1).

$$J = \frac{v}{At} \quad (1)$$

Where, v is permeate volume in m^3, t is time in second, A is membrane area in m^2 and J is calculated permeate flux in $m^3 m^{-2} s^{-1}$.

2.5. Sample analysis

The physico-chemical characteristics, such as, bulk density, porosity, BET surface area (Coulter, SA3100, Germany), particle size (Mastersizer, APA 2000, UK), moisture content and point of zero charge (Beckman Coulter, Delsa™ Nano C, Germany) were measured. The characteristic values of the biosorbent are tabulated in Table 1. The pH of solution were measured using a pH meter (Eutech Instruments, pH 700, India). The functional groups present in biosorbent were identified by FTIR transmittance spectra of native as well as Pb ion loaded adsorbent (Shimadzu, IR Affinity-1 FTIR spectrometer, Japan). The elemental analysis of native and Pb ion loaded adsorbent were done by FESEM-EDX (Zeiss, Sigma, USA). The Pb ion concentration in solution was measured using atomic adsorption spectrometer (AAS) (Varian, AA240AS, Netherlands).

2.6. Experimental setup

A rectangular cross flow membrane module setup was fabricated with the help of IIT Guwahati mechanical work shop, details of its parts are tabulated in Table S1. Biosorption process was combined with membrane process in two scheme; (1) Biosorption process followed by membrane process