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Original article

Using pretreated chestnut endothelium to adsorb lead and cadmium ions from water

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ABSTRACT

The nature chestnut endothelium, as waster source from chestnut (*Castaneamollissima*) has pigment effecting the process of adsorbing heavy metalions, and the decolorized endothelium has low adsorption capacity. In order to raise the adsorption capacity of heavy metal ions, the discolor endothelium was pretreated by acidic formaldehyde, cis-butenedioic acid and irradiation. Thermodynamic and kinetics model was fitted to the adsorption of Pb (II) and Cd(II) ions onto modified chestnut endothelium by cis-butenedioic acid. Three independent variables including pH, adsorption time and contact temperature were selected as affecting factors to Response Surface. The modified experiment results showed adsorption rate of Pb(II) and Cd(II) ions on the chestnut endothelium modified by 0.5 mol/L cis-butenedioic acid was higher than other modified methods. Thermodynamic and kinetics model was fitted with Langmuir and Pseudo-second-order kinetic model, respectively. 59.23 °C of the adsorption temperature, the 5.72 h of adsorption time and the 6.16 of pH are the optimized conditions of the adsorption rate of Pb^{2+} on modified chestnut endothelium. 55.93 °C of the adsorption temperature, the 4.43 h of adsorption time and the 6.06 of pH are the optimized adsorption conditions of Cd^{2+}. Under the optimized condition, the experiment value of the adsorption of Pb^{2+} and Cd^{2+} was 99.76% and 98.90%, respectively, which are close to the predicted value. The FTIR indicated that C—O, O—H and C—H involved in the adsorption process of Pb^{2+} and Cd^{2+}.

1. Introduction

Along with the industrial activity and economic development, the heavy metal ions which was not biodegradable toxic chemicals pollutant, pose directly threat to human health and environment (Liu et al., 2008; Nema et al., 2017), because of toxicity, increased accumulation on the food chain and ecosystem (Lu et al., 2015a, b; Xiao et al., 2015).

Association of cadmium ions with enzymes can reduce their activities, influence fat and protein metabolism or digestion, easily induce heart cerebrovascular disease and high blood pressure. Cadmium ion also can exchange the calcium in calcium phosphate in human bone, and lead to bone osteoporosis due to the lack of calcium in the bone. Moreover, Cadmium is accumulated in the kidneys, liver and bone marrow, replace zinc ion in specific enzymes, and make the body prone to renal artery, sex atrophy, diabetes and other diseases (Du et al., 2015). Lead ion is also toxic to the human body. The human bone will store about 95% of lead, in the form of phosphate salts, and a small amount of lead will exist in the liver, kidney, spleen, lung, heart, brain, bones and muscles. Hence the toxicity of Pb ions causes damage to liver, kidney, mental retardation and infertility and abnormalities in pregnant women (Singh et al., 2008; Singh et al., 2007; Ismail and Harons, 2017).

Among the techniques applied to the removal of heavy metal from wastewater are chemical precipitation, chemical oxidation or reduction, electrochemical, and membrane separation (Huang et al., 2007; Amuda et al., 2007). However, these methods require high capital investment and cost of the equipment maintenance fee (Li et al., 2013), as well as creating sludge disposal problem, which contains a lot of by-products (Aziz et al., 2008; Wong,

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2017). Therefore, most of the recent researches to treatment of heavy metals in aqueous solutions was interested in much more effective low-cost adsorbents made from nature materials (Caner et al., 2015; Şenet al., 2015; Gao, et al., 2017), such as biosorption, for removing heavy metals from wastewaters (Rojas et al., 2015; Hasan and Hanafiah, 2017). This, method use biomass which are the byproducts of agriculture, forestry and forestry waste to adsorb metal ions. There are many researches in which different waste bio-adsorbents in the world are used as adsorbent. For example, Lu (Lu et al., 2015a,b) used rice straw modified by fermentation and simple chemical treatment to remove chromium (III) from industrial wastewater. Yurtseverinvestigated the effect of pH of solution, contact temperature and initial concentration of Pb^{2+} on Pb (II) biosorption onto modified quebracho tannin resin (Yurtsever and Şengil, 2009). Humelnicustudied the sorption performances of two kinds of by-products (soy bran and mustard husk) resulted from the agriculture (Humelnicu et al., 2015). Dahiya used pre-treated crab and arca shell as adsorbent of lead and copper from aqueous solutions (Dahiyaet al., 2008). Zhang studied on the kinetics and thermodynamics of Pb^{2+}, Cu^{2+}, Cr^{6+} and Cd^{2+} adsorption onto peanut hull (Zhang et al., 2010).

Chinese chestnut (*Castaneamollissima*) is a member of the familyFagaceae, and a species ofchestnutnative toTaiwan, Korea and China. It grows widely in Hebei, Liaoning, Shandong, Hunan, Hubei, Jiangxi, Fujian, Anhui, *etc*. The total annual production of 825,000 tons, accounted for over 75% of the total world Chinese chestnut. Traditional products of chestnut are sugar Fried chestnut, beverage, canned, frozen, and wine (Yang et al., 2007). In the process of those products, the endothelium after taking the production of chestnut has become the agriculture and forestry waste. The main component in the Chestnut endothelium is the lignin and cellulose, which account for about 80% of total weight. The remaining 20% consists of pentose, tannins, flavonoids, organic acids, phenols, phytosterol, glycosides and lactones. Chinese chestnut endothelium as a biological adsorbent was applied to the adsorption of heavy metals in water environment has been studied by some researches (Qi et al., 2009; Wang, 2009; Wu and Huang, 2010; Ding et al., 2010; Samad et al., 2017).

However, when chestnut endothelium was used as the adsorbent of the heavy metal ions, the pigment in the endothelium are easily dissolved in water, causing the change of the colors and limiting the reuse of aqueous solution after the removal of heavy metals. Moreover, the pigment can react with heavy metal ions and cause the precipitation of heavy metal ions. Therefore, removed the pigment material, which could be a nature pigment added into the food, are required before the chestnut endothelium is used as biological adsorbent. However, the real adsorption rate of Pb and Cd ions onto decolorized endothelium did not reach 80% and was relatively low (Zhang et al., 2016).

Vázquez (Vázquez et al., 2009) uses acid formaldehyde pretreated chestnut endothelium asadsorbent tooptimize the removal of Pb(II), Cu(II) and Zn(II) from aqueous solutions. The maximumadsorption capacity of pre-treated chestnut shell was obtained for Pb (II) ions, 8.5 $mg \cdot g^{-1}$, and the order of cation affinity was $Pb^{2+} > Cu^{2+} > Zn^{2+}$. Vázquez (Vázquezet al., 2012) also studied that chestnut endothelium was pretreated by an alkaline solution to increase its adsorption capacity, and the influence of the concentration of NaOH and contact time on Cd (II) ion adsorption. The highest adsorption capacity of Cd(II) ion was 9.9 $mg \cdot g^{-1}$.

The objective of this research was to modify the decolorized chestnut endothelium to improve the adsorption rate and sorption capacity of Pb(II) and Cd (II) ions from aqueous solution. The effects of the different experimental conditions on the chestnut endothelium efficiency for removing of Pb(II) and Cd (II) ions were also investigated. The isotherms and dynamics were studied in the sorption process. This work increased the sorption capacity of Pb(II) and Cd (II)ions, which reached 52.91 mg/g and 51.28 mg/ respectively. It is important for us to reuse the waster of chestnut shell and apply the modified endothelium to remove the heavy metal pollution from the environment.

2. Materials and methods

2.1. Materials

The chestnut (from the market of Baoding, China) was peeled manually, and the endothelium was washed thoroughly with distilled water. It was dried in the air at the room temperature. Then it was groundedand sieved through with 40 mesh size. Finally, the natural chestnut endothelium was obtained and named as NO. 1 material.

The discolor process was performed in a 1 L glass Pyrex glass reactor with mechanical stirring. 5 g of endothelium power was immersed in 400 mL sodium Hydroxide solution (0.1 M) at 70 °C for 5 h, and then the suspension was filtered by filter paper. The solid residue was discolored twice more, and then was rinsed by distilled water until the pH of wash liquor was the same as that of the distilled water. Finally, the chestnut endothelium was dried in the vacuum dryer and the Decolorized chestnut endothelium (named as NO. 2) was obtained (Zhao et al., 2015).

2.2. Instrument and equipment

The SZ-93 automatic double distill Baporizer for the purification of water was made by Shanghai Yarong Biochemical Instrument Plant, China. Electronic analytical balanceAR423CN and pH meter (Ohaus Instrument Co. Ltd, Shanghai, China). Super-heated water bathHH-601 (Hengfeng Instrument Co.Ltd, Jintan, Jiangsu, China). TAS-990-supper atomic adsorption spectrophotometer (Beijing Purkinje General Instrument Corporation, China).^{60}Co-γ radiant Point (Heliyuan LID. CO. of Baoding, Hebei, China). FTS3000 FTIR Spectrometer (BIORAD Corporation).

2.3. Reagents

Sodium hydroxide, hydrogen nitrate, sulfuricacid, perchloricacid, formaldehyde, cis-butene dioicacid, lead nitrate and cadmium nitrate were all of analytic grade from Tianjin Chemical Co, China.1000 $mg \cdot L^{-1}$ Lead standard solution and cadmium standard solution were obtaining from Nation Institute Metrology, China. All the water was double distilled one made from the double distillation of the ion-free water.

2.4. Modification chestnut endothelium

2.4.1. Modified by Cis-butenedioic acid and formaldehyde acid

The discolored chestnut endothelium was put into flasks, and 0.5 mol/Lcis-butenedioic acid or formaldehyde acid (0.1 M sulfuric acid:37% formaldehyde = 4:1) were also added individually into the flash with solid-liquid ratio of 1:20 and 2:5 (g:mL), respectively. The solutions in the flask were placed into the water bath at 50 °C for 2 h and then were filtered. The solid fractions were collected washed with distilled water to pH > 4 and was dried in vacuum drier for 6 h, at 60 °C, 0.08 MPa. The endothelium modified by cis-butenedioic acid and formaldehyde acid were named as NO. 3 and NO. 4, respectively.

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