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Converting spent battery anode waste into a porous biocomposite with high Pb(II) ion capture capacity from solution

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ABSTRACT

Taking advantage of the low-cost carbon and manganese sources of the spent battery anode waste (SB), porous biocomposites were synthesized by co-pyrolyzing of SB with sawdust. Synthesis method was optimized, and potential biocomposite application in the aspect of Pb(II) removal in the wastewater was investigated and the related possible mechanism of Pb(II) removal was discussed. Composite derived at 750 °C with 10% SB loading amount had high Pb(II) adsorption capacity with negligible influence of ion strength. The maximum Pb(II) adsorption capacity was around 183.82 mg/g at 298 K and pH 4, and the increase of the temperature favored Pb(II) adsorption. Pb(II) adsorption behavior fitted the intraparticle diffusion model and Redlich-Pearson model. Pb(II) removal was governed by precipitation, complexation and C_{π}–Pb(II) bond interaction combined processes. Co-pyrolysis of sawdust with SB not only explores an approach for the SB utilization but also provides an excellent biocomposite for solution Pb(II) ion adsorptive removal.

1. Introduction

Alkaline batteries are the most widely used disposable primary batteries in various types of electric and electronic gadgets of modern life. The improvement in the quality of life and general lifestyle in China resulted in more than 0.1 million metric tons of spent batteries waste disposal into the environment (Zeng et al., 2017). Recovering and recycling spent batteries waste is now a globally sustainable development strategy since such a large-scale production of waste is becoming an urgent environmental threat (Zeng et al., 2015a). The pyro- and hydro-metallurgical methods are most widely used approaches for recycling the valuable resources from spent batteries waste. Many metal and cathode materials had been successfully recovered via acid leaching or solvent extraction (Biswas et al., 2016; Swain et al., 2007; Zeng et al., 2015b). However, recovering and exploring the utilization of spent batteries anode waste are seldom investigated till now.

Spent batteries anode waste (SB) are mainly composed of carbon-based material residues featuring with large quantity, relatively pure components, superior carbon matrix structures, etc. It was considered as the potential and suitable alternatives for preparation of carbon-based adsorbent in the process of wastewater treatment (Ali et al., 2017; Demiral and Güngör, 2016; Zhang et al., 2016b). And the novel carbon-based magnesium hydroxide nanoparticle derived from spent Li-ion battery anode with phosphate adsorption capacity around 588.4 mg/g had been successfully synthesized (Zhang et al., 2016a). Similar to Li-ion battery, alkaline Zn–MnO_2 battery is also the most popular portable battery in the market which covers ~60% of global primary battery production (Buzatu et al., 2013). After the Zn and Mn recovery, exploring the utilization of the resulted SB of the Zn–MnO_2 battery is a major issue for the sustainable development strategy.

Nowadays, water pollution has become a severe global issue due to the large amount of toxic heavy metal containing effluents discharged into the environment (Li et al., 2018a). Lead (Pb) is one of non-biodegradable contaminants in discharged effluents characterized by high toxicity, easily accumulating through food chains

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and causing serious diseases and disorders even at low concentration (Goyer, 1993). Removal of Pb(II) from the water is a burning issue to meet stringent environmental quality standards. Currently, various methods could be adopted for Pb(II) removal in industrial effluents to a permissible level (El-Korashy et al., 2016). Adsorption treatment has attracted considerable attention, owing to its comparative usefulness and cost-effectiveness in practice (Adebisi et al., 2017; Elwakeel et al., 2018; Li et al., 2018b). While, the traditional activated carbon adsorbent often shows less than 30 mg/g Pb(II) adsorption capacity (Kolodynska et al., 2017) and the graphene-based adsorbents are relatively expensive in use (Ren et al., 2012). Recently, some studies showed low-cost engineered carbon composite derived from treatment of biomass or biochar with $KMnO_4$ solution exhibited acceptable adsorption capacities for Pb(II), Cd(II), As(V), and Cu(II), etc. (Wang et al., 2015a, 2015b; Yu et al., 2017). The improvement of adsorption capacity is attributed to the presence of Mn oxides particles on the carbon surfaces of biochar, which provides more active sites for contaminant adsorption from the solution.

In this sense, fabricating Mn-containing compounds on the recovered carbon materials from spent alkaline Zn–MnO_2 battery anode theoretically has great potential to strongly adsorb Pb(II) from wastewater. Considering the features of carbon materials, products harvested from the SB probably carried minor amounts of Mn on the carbon matrix after recycling operation. The residual Mn in SB perhaps is an alternative low-cost Mn source prior to traditional $KMnO_4$ salt in theory. Moreover, using the Mn-containing SB waste as one kind of Mn-containing sources not only can explore the utilization of spent batteries anode waste, but also can provide a special adsorbent for wastewater treatment. While, this assumption is still uncovered, fewer reports are conducting on the adsorbent exploration from recycled spent alkaline Zn–MnO_2 battery anode waste, and the potential for Pb(II) adsorptive removal from aqueous medium is kept unknown.

Herein, SB based porous biocomposites (SBC) were synthesized by co-pyrolyzing of SB with sawdust for adsorption of Pb(II) in aqueous solutions. Co-pyrolysis condition was optimized and the structure of the as-prepared SBC sample was characterized. Factors influencing Pb(II) removal efficiency were investigated, and possible mechanism involved in the Pb(II) adsorption by SBC adsorbent was discussed.

2. Materials and methods

2.1. Materials

Sawdust was collected from a local drum factory near the north campus of Northwest A&F University, Yangling, China. The biomass material was washed by tap and deionized water (DI, 18.2 MΩ) to minimize the presence of dust followed by oven-drying at 105 °C for 6 h. The dry sawdust was crushed to size <0.12 mm before use. Commercial, cylindrical Nanfu brand (Nanping, China) spent Zn–MnO_2 batteries were used as primary battery waste materials. Before use, battery steel crusts were dismantled; anodes and cathodes were manually uncurled and separated. The black anodes powder sample analysis showed that the major metallic constituents were Mn (35.4 ± 0.3%), Zn (11.2 ± 0.2%) and Fe (2.5 ± 0.1%). The purification of SB was conducted by using the leaching procedure recommended by Biswas et al. (2016). In brief, 20 g of SB powder was firstly immersed into a 1000 mL solution containing 2 g glucose and 2 mol sulfuric acid at 100 °C under magnetic stirring for 1 h. Then the black SB powder were filtered off, washed thoroughly with DI water three times to remove residual leaching reagents and other impurities, and dried in an oven at 105 °C for 6 h before use. All chemicals, including sodium hydroxide, nitric acid, hydrogen peroxide, ethylene diamine tetraacetic acid sodium salt (Na_2EDTA), and glucose of analytical grade were purchased from Xilong Chem. Co., Ltd. (Chengdu, China) and used as received. Working solution of Pb(II) was prepared by diluting 1000 mg/L standard stock solution (Fisher Scientific). All chemical solutions were prepared using DI water in the experiment.

2.2. Preparation of the porous biocomposites

The SB based porous biocomposites (SBC) were synthesized by a co-pyrolysis method. Simply, a desired amount of dehydrated SB powder was first thoroughly mixed with dry sawdust. Then the mixtures were heated in a muffle furnace at 350, 550 and 750 °C for 1 h under N_2 flow of 200 mL/min, respectively (Jeong et al., 2015). The resulted samples were then washed with DI water, dried in the oven at 105 °C and ground into fine powder. Samples with and without SB powder were prepared in this study. For the convenience of discussion, BC refers to the prepared carbon material without SB, while xSBCy refers to SB based porous biocomposites where *x* represents the specific amount of SB (%), and *y* denotes the pyrolysis temperature. All the obtained samples were stored in a desiccator for further experiments.

2.3. Biocomposites characterization

Elemental C, H, O, and N analysis were performed on a CHON elemental analyzer (Elementar Analysensysteme GmbH, Germany) via high-temperature combustion of composite samples. Major metallic constituents of the composites were determined by inductively coupled plasma optical emission spectroscopy (ICP-OES, Varian, USA) after digestion of samples with concentrated HNO_3 and H_2O_2 at about 150 °C. The BET surface area (S_{BET}), the total pore volume (V_{tot}) and the average pore size (*AVP*) of the samples were evaluated based on N_2 adsorption at 77 K using a V-Sorb 2800P analyzer (App-one, China). Surface functionalities of samples were examined by a Fourier-transform infrared spectrometer (FT–IR) (Vetex70, Bruker Corp, Germany) with KBr slice in recording range of 400–4000 cm^{-1}. Surface morphology of samples were recorded by using a scanning electron microscope (SEM, JEOL 6335F, Japan) equipped with an energy dispersive X-ray spectrometer (EDX, Quantax70, Bruker Corp, Germany). X-ray diffraction (XRD) patterns were obtained by using a powder diffractometer (Bruker D8 advance, Bruker AXS, USA) with a Cu Kα source. X–ray photoelectron spectroscopy (XPS) data were collected using an Axis Ultra DLD X–ray photoelectron spectrometer equipped with an Al Kα X-ray source (1486.6 eV).

2.4. Batch adsorption experiments

Batch adsorption experiments were undertaken to study the Pb(II) adsorption onto biocomposites. In optimizing the Pb(II) adsorption performance of the as-prepared biocomposites, 0.05 g samples were mixed with a series of 60 mL 100 mg/L Pb(II) solutions (pH 4.0, which was optimized in previous study) in centrifuge tubes on a shaker platform operating at 120 rpm under room temperature (25 °C). The pH-dependent adsorption experiments were performed over an initial pH range of 1.5–5.0 adjusted by 0.1 M HNO_3 and NaOH solution. Adsorption isotherms were conducted with varying initial concentrations of Pb(II) (10–500 mg/L, pH 4.0). The effect of ion strength on Pb(II) adsorption was investigated by adjusting the Pb(II) solution with $NaNO_3$ concentration varied from 0 mmol/L to 100 mmol/L. For investigation of the Pb(II) adsorption kinetics, about 600 mL Pb(II) solution (pH 4.0, 100 mg/L) was mixing with 0.5 g biocomposite in a plastic bottle. The mixtures were magnetically stirred at 120 rpm at room temperature for