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Journal of Environmental Chemical Engineering



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

# Selective toxic metal uptake using an e-waste-based novel sorbent-Single, binary and ternary systems

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#### ARTICLE INFO

#### ABSTRACT

Article history: Received 22 October 2013 Accepted 4 January 2014

Keywords: Toxic metal removal Selective adsorption Wastewater treatment Waste management E-waste Since the disposal of printed circuit boards pose worldwide, dire environmental threats for nations, it is imperative to tackle this serious problem by adequate solutions. The modification of the non-metallic fraction of this waste (NMP) results in the development of a nano-structured material as well as proper functionalization, which renders it a potentially attractive material to be employed as an adsorbent to capture toxic metals from effluents. The removal efficiency of the modified nano-porous material (A-NMP) for copper, lead and zinc in single-component solutions have been measured 2.9 mmol/g, 3.5 mmol/g and 2.0 mmol/g, respectively which is ranked much higher than its industrial counterparts. Furthermore, partial selectivity was observed in the binary solution of copper and lead, whereas there was an absolute selectivity in zinc-containing binary solutions in favor of the other metal, i.e. lead or copper. The ternary system demonstrated a similar trend to the binary system.

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#### Introduction

Toxic metals are well suited to be used extensively in various industries including microelectronics, jewelry, electroplating, etc. due to their specific characteristic properties [1]. The high demand for metal and its mounting consumption together with depleted global reserves have obliged the industries to seek for an efficient, cost-effective and systematic approach to recover and reuse them instead of continuous ever-increasing extraction of metal minerals from the ore deposits [2]. In addition, escalating environmental implications linked to the toxicity of metals and their negative impacts on soil and groundwater contamination and hence public health risks pose the foremost global concerns which must be meticulously tackled [3]. Accordingly, selective uptake of toxic metals from waste streams is a key task ahead which not only contributes to ecological protection but also to a sustainable development with social impacts and economic rewards.

A diversified array of conventional technologies applied in the industry to remove toxic metals from wastewater encompass chemical precipitation [4], flotation [5], membrane [6], electrochemical treatment [7], reverse osmosis [8] and solvent extraction [9]. Nevertheless these traditional techniques present their respective drawbacks, namely soaring cost, high energy requirement, as well as toxic sludge formation which needs to be further dewatered, disposed and detoxified rendering it almost as problematic as the treatment of the original wastewater.

With the progression of technology, adsorption has gained momentum as a viable alternative to the traditional technologies for the removal of the toxic metals from the effluents in the light of its convenience, ease of operation and simplicity of design [10]. Activated carbons have been the focus of attention for this purpose because of their high uptake capacities owing to their porous structures and various surface functional groups. The everincreasing demand for these materials necessitates the exploration for new sources of low-cost, sustainable precursors [11]. During the last two decade, many studies have been pursued by the researchers on the use of waste agricultural products such as sugarcane bagasse [12], olive stone [13], wheat straw [14], rice husk [15], sawdust [16], palm shell [17], date pits [18], peanut hull [19], almond shell [20], etc. as precursors for the production of activated carbons and their application for wastewater treatment. However, the focus of most of these researches has been the removal of single metals from the aqueous solutions rather than selective adsorption of the metals for recycling purposes.

Zeolites as naturally occurring hydrated aluminosilicate minerals are another class of materials that have received much attention as adsorbents due to their unique structures consisting of three-dimensional frameworks of SiO<sub>4</sub> and AlO<sub>4</sub> tetrahedra with a well-defined network of pores. The substitution of silicon atoms in the structure of the zeolites by aluminum atoms results in the formation of a net negative charge in the lattice which is

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neutralized with a small cation such as sodium or calcium. The existence of such cations allows the removal of undesirable toxic metal ions from industrial effluents by an ion-exchange mechanism [21].

Reuse of zeolitic waste materials as adsorbents is still in its infancy. Sočo et al. have studied the possibility of toxic metal adsorption on fly ash [22]. Waste Sandstone Cake has been converted to zeolite by Takaaki et al. [23] and has been applied for the removal of Strontium. Similarly, Ibraheem et al. [24] synthesized ZSM-5 zeolite from rice husk for lead ion adsorption. Some other researchers have reported the synthesis of zeolite from industrial wastes, e.g. paper sludge ash, municipal incinerated ash, and waste perlite [25–27].

Although it is a common knowledge that printed circuit boards (PCBs) comprise of aluminosilicate materials rendering it theoretically an ideal tool for adsorption principles, to the best knowledge of the authors, no attempt has been made to manipulate obsolete waste printed circuit boards (PCB) to be reused as potential toxic metal adsorbents. Hence, the aim of this study is to explore the viability of converting this low-value, obsolete waste material to an innovative value-added material to immobilize the toxic metal ions present in the wastewater.

#### Materials and methods

#### Materials

Potassium hydroxide (KOH, >85%) was purchased from Sigma-Aldrich and used as received.

Virgin non-metallic fraction of printed circuit boards (NMP) was acquired from a local company in Hong Kong.

#### Production of activated material

Dried precursor (NMP) was impregnated with 1 M KOH solution for 3 h at room temperature in air. The slurry was then transferred to the reactor and activated at 250 °C at a rate of 5 °C min<sup>-1</sup> for 3 h under continuous nitrogen flow. After the activation process, the reactor was cooled down to room temperature in a nitrogen atmosphere. The resultant material was washed with hot and cold water for several hours to ensure the removal of ions and inorganics. After washing, activated sample was dried at 110 °C for 24 h.

#### Characterization of activated material

The BET surface area was determined from the nitrogen adsorption isotherm by using Brunauer–Emmett–Teller (BET) equation. The BET surface area measurements were obtained from nitrogen adsorption isotherms at 77 K using a Quantachrome Autosorb-I Surface Area Analyzer. Automatic equipment software (AS1Win 1.5) was used to obtain the nitrogen adsorption isotherms. To determine the micropore volumes and the external surface area, the *t*-plot approach was applied. Fourier transform infrared (FTIR) spectra of the original and activated material were obtained using a spectrophotometer (FTS 6000) in order to qualitatively identify the chemical functionality of activated materials. Pellets were prepared by mixing 1 mg of dried sample with 100 mg of KBr (Merck, for spectroscopy). The surface structures of the treated and untreated materials were further investigated by X-ray photoelectron spectroscopy (XPS, PHI5600).

#### Aqueous adsorption characteristics

The adsorption tests were performed by batch technique at 20 °C. For isothermal studies in a single component system, a series

of 50 mL metal solutions at different initial concentrations in plastic bottles were prepared and each bottle was loaded with the target adsorbent at a mass loading of 1 g/L. The solution pH was adjusted to 4 by 0.1 M HNO<sub>3</sub> and NaOH solutions. The bottles were then agitated in an orbital shaker at 120 rpm and liquid samples were taken out at specified time intervals for analysis and filtered in order to avoid further adsorption. The metal concentrations were determined by using an Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES, Optima 7300 DV, Perkin Elmer).

The effect of pH on the adsorption of toxic metal ions onto the adsorbents was also conducted by varying the pH level from 1.5 to 6.

For co-adsorption in binary systems of  $Pb^{2+}/Cu^{2+}$ ,  $Pb^{2+}/Zn^{2+}$  and  $Cu^{2+}/Zn^{2+}$  and the ternary system of  $Pb^{2+}/Zn^{2+}/Cu^{2+}$ , 1 g/L of adsorbent was mixed with 50 mL equimolar concentrations of respective metal solutions. The procedure for testing and analysis was the same as described above.

#### **Results and discussion**

#### Elemental composition

One of the preliminary steps to identify the mechanism underlying the activation process is to look into the elemental composition of the material before and after the activation process. CHNS elemental analyses for the treated and untreated materials demonstrated that the carbon content of the original waste material (NMP) is around 20 wt%, which is burnt off after the activation process despite the low processing temperature probably due to the catalyzing effect of potassium and calcium [28,29] (see Table S1).

Moreover, X-ray fluorescence spectrometry (XRF) indicated that the major elements present in NMP include silicon, aluminum, and calcium which are believed to originate from calcium aluminosilicate, one of the major constituents of printed circuit boards. Table S2 suggests that this composition does not change significantly after the activation process which could be related to the high thermal stability of aluminosilicate materials.

#### Surface characterization

In order to track the changes in the functionalities of the material before and after activation process, Fourier transform infrared spectroscopy (FTIR) and X-ray photoelectron spectroscopy (XPS) measurements have been performed on the original (NMP) and treated material (A-NMP).



Fig. 1. FTIR spectra of the treated and virgin materials.

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