Synergistic effect in the simultaneous removal of binary cobalt–nickel heavy metals from effluents by a novel e-waste-derived material

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HIGHLIGHTS

- Production of a novel activated adsorbent material from the non-metallic component of PCB e-waste.
- High performance for the removal of metal ions both in single- and binary-component systems.
- Higher cobalt and nickel removal capability in comparison with the industrial adsorbents.
- Synergistic effect of cobalt and nickel in binary-component systems.
- Best fit equilibrium isotherm modeling in single and binary component systems.

ARTICLE INFO

ABSTRACT

Simultaneous removal of heavy metals coupled with high adsorption capacity of an adsorbent material is an ultimate target in industry for the purification of wastewater. A novel adsorbent prepared from an electronic waste-based material has been used for this purpose. The results exhibit very high removal capacities of the adsorbent for both cobalt and nickel in the single-component systems. These capacities have been compared with those of three widely-used industrial adsorbents and it has been shown that the removal capability of this novel material is well above those of the industrial ones. Moreover, the effect of pH has also been explored and it has been shown that the pH value has a drastic effect on the metal removal capacity at pH values lower than 3. Furthermore, a synergistic effect of two metals has been observed and studied in the binary metal system. The results indicate that not only the simultaneous presence of the two metals does not reduce the adsorption capacity, but also enhances their removal from the effluent.

Moreover, the adsorption equilibrium modeling for the single and binary systems have been presented and it has been inferred that the Langmuir-type models can fit the single component experimental data, while the binary system can be fitted only by the modified extended Freundlich model.

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1. Introduction

Cobalt and nickel are two widely-used heavy metals which are employed in various industries such as refining industry, textile and paint industry, foundries and metal finishing industry. Therefore, the challenge associated with the treatment of nickel- and cobalt-containing wastewater discharged from these industries is important due to their ecological impact and human health threat [1].

There are numerous methods for the removal of these heavy metal ions from wastewater streams including chemical precipitation [2], reverse osmosis [3], filtration methods [4], electrocoagulation technique [5] and adsorption [6–8]. Among all these techniques, adsorption has been found to be a promising method that does not have the disadvantages of others such as low efficiency and high cost.

At present, low cost adsorbents made up of carbonaceous waste materials such as bamboo [9], peat [10], tyre [11], bagasse [12] and bone char [13] have been of great interest for the purpose of pollution removal from wastewater. However, there are two major issues with these adsorbents, namely their low yield and very high processing temperatures required for their activation.

Zeolites are another class of adsorbents which are being used in industry for this purpose. The structure of these materials makes them specifically attractive for adsorption owing to their highly ordered nano-structured framework [14]. The synthesis process for these materials with well-defined network of pores is a very tedious task leading to the high cost of these materials. According to Hui et al. the capital cost for the zeolitic adsorbents can be much higher than the activated carbons [15].

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A few studies investigated the use of waste zeolite-structured materials to adsorb heavy metal ions from effluents. Qiu and Zheng used fly ash to remove heavy metals from wastewater [16]. A zeolite-like material extracted from a waste in pumice mining was obtained by Catalfamo et al. [17]. Nevertheless, to the best knowledge of the authors, no literature has aimed at the encapsulation of heavy metal ions in wastewater streams onto the non-metallic fraction of waste printed circuit board materials (nonmetallic powder, NMP), made up of aluminosilicate materials used in the present study.

The focus of this paper is the simultaneous uptake of nickel and cobalt at high concentrations either as a single metal or as co-existing ions onto this novel adsorbent material after the modification process and also the investigation of their synergistic effect. The potential of this work is not only in wastewater treatment, but also resource recycling resulting in an issue of global concern becoming a value-added product opportunity.

2. Experimental

2.1. Materials

Micron-size e-waste raw material (non-metallic powder, NMP) was acquired from a local company in Hong Kong and was used as received. Potassium hydroxide (KOH, >85%), and nitrate salts of cobalt (II) and nickel (II) were purchased from Sigma–Aldrich Company, UK.

2.2. Activated material preparation

A chemical activation method using potassium hydroxide as the activating agent was used for this purpose. The NMP e-waste precursor was impregnated with the activating agent with an impregnation ratio of 2 with continuous stirring for 3 h at room temperature where impregnation ratio is defined as the weight ratio of the activating agent to the precursor. Subsequently, the resulting slurry was activated in a 18 L muffle furnace (AAF 11/18, Carbolite, UK) under flowing nitrogen (purity 99.99%+) at 250 °C at a rate of 5 °C min⁻¹ for 3 h. Afterwards, the furnace was cooled down to room temperature in a nitrogen atmosphere. The resultant material was then washed with hot and cold water followed by drying at 110 °C for 24 h and stored in a desiccator for later experimental use.

2.3. Porosity and surface area determination

The surface areas of the virgin and activated material were determined by the nitrogen sorption method using Quantachrome Autosorb-I Surface Area Analyzer, USA. The dried samples were outgassed at 423 K for 6 h followed by nitrogen sorption at 77 K. The total surface area and the micropore volume were determined by the Brunauer–Emmet–Teller (BET) and t-plot equations, respectively. The total pore volume (i.e. sum of micro- and mesopore volume) is defined as the volume of nitrogen adsorbed at $p/p_0 = 0.98$. The mesopore volume is the difference between the total and micropore volumes.

2.4. X-ray photoelectronic spectroscopy (XPS)

X-ray photoelectron data were acquired with an XPS-PHI5600 system, USA with a monochromatic Al Kα source (excitation energy, $h\upsilon = 1486.6$ eV) at a voltage of 10 kV and a current of 15 mA. Low resolution spectrum for the whole range was acquired at 70 eV pass energy, whereas high resolution narrow range spectra for the elements of interest were acquired at 20 eV pass energy.

The spectra were corrected using C (1s) at 284.6 eV binding energy as reference.

2.5. Fourier transform infrared spectroscopy (FTIR)

FTIR spectra were recorded using Perkin Elmer spectrometer (Spectrum GX, USA), by KBr pelletization method from 400 to 4000 cm⁻¹. 3 mg of sample was accurately weighed and ground with 300 mg of spectral grade KBr. After thorough mixing using mortar and pestle, the contents were made into a pellet form by applying 7 tons of pressure. The obtained pellet was subjected to FTIR quantitative analysis.

2.6. Aqueous adsorption characteristics

Cobalt and nickel metals were used as adsorbates in single- and multi-component systems to evaluate the adsorption capacity of the prepared activated material.

In a single component system, different concentrations of solutions containing a metal ion were prepared by dissolving appropriate amount of the specific metal in distilled water. Adsorption isotherms were derived from the batch experimental data. Test solutions (50 mL) of various concentrations (ranging between 0.5 and 5 M) with a pH level of 4 were added to the adsorbent (0.05 g) and the suspensions were shaken at 298 K for an equilibrium time determined by preliminary contact time studies (see Fig. 1). The filtrates were analyzed for the residual target metal concentration using an inductively coupled plasma atomic emission spectroscopy (ICP-AES, Optima 7300 DV, Perkin–Elmer, USA). The amount of the metal adsorbed was calculated using the following equation:

$$q_e = \frac{[c_o - c_e]V}{M} \tag{1}$$

where q_e is the amount of metal ion adsorbed per unit amount of the adsorbent (mg/g); C_0 and C_e denote the concentrations of metal ion in the initial solution (mg/L) and after adsorption respectively; V is the volume of the adsorption medium (L); and M is the amount of the adsorbent (g).

A similar adsorption technique was employed for the binary-component system. Different concentrations of equimolar Ni and Co solutions (ranging from 0.5 M to 5 M) at a pH level of 4 were prepared for binary-component adsorption purposes. The rest of the test procedures and the analyses were the same as the single-component adsorption.

Notably, all the experiments were repeated at least three times in order to ensure the reliability of the acquired data.
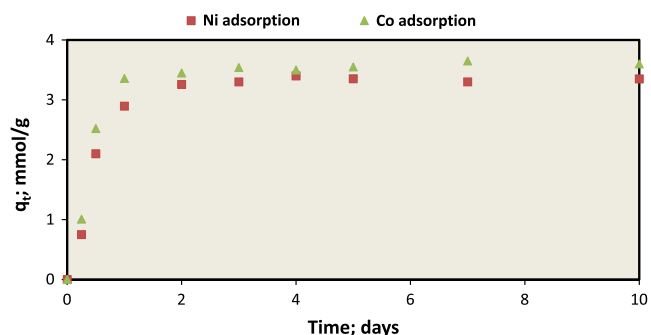

Fig. 1. Equilibrium contact time for 4 mM nickel and cobalt solutions at a pH level of 4.