



Lithium recovery from spent Li-ion batteries using coconut shell activated carbon



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ABSTRACT

Lithium is one of scarce natural resources in the world that need to be preserve. One of the way in preserving the resource is by recovery the rich source of the lithium such as in the spent batteries. It is necessary to develop a recovery method which is efficient and low-cost to be able to recover the lithium in an economic scale. In this study, low-cost activated carbon (AC) from coconut shell charcoal was prepared by chemical and physical activation methods and tested for Li removal from Co, Mn, and Ni ions in semi-continuous columns adsorption experiments. The maximum surface area is 365 m²/g with the total pore volume is 0.148 cm³/g that can be produced by physical activation at 800 °C. In the same activation temperature, activation using KOH has larger ratio of micropore volume than physical activation. Then, the adsorption capacity and selectivity of metal ions were investigated. A very low adsorption capacity of AC for Li ions in batch adsorption mode provides an advantage in column applications for separating Li from other metal ions. The AC sample with chemical activation provided better separation than the samples with physical activation in the column adsorption method. During a certain period of early adsorption (lag time), solution collected from the column outlet was found to be rich in Li due to the fast travel time of this light element, while the other heavier metal ions were mostly retained in the AC bed. The maximum lag time is 97.3 min with AC by KOH activation at 750 °C.

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1. Introduction

The demand for Li-ion batteries significantly increases and dominates the battery share, especially following the increase of electric vehicles and electronic products in the market (Meshram et al., 2015), due to their advantages of high energy density, large working temperature range, long circle-life, low self-discharging rate, and high working-voltage (Wang et al., 2016; Jeong et al., 2015; Aziz et al., 2016). Li-ion batteries are mainly composed of cathode (Li metal oxide), anode (graphite), electrolyte, separator (PVDF) and metal casing (Barik et al., 2016). However, as the consequence of this large implementation of Li-ion batteries, huge amounts of spent Li-ion batteries are generated. On the other hand, Li is well known as a rare-earth metal having high economic value, and available mainly in several specific regions, including Argentina, Bolivia, and Chile. Therefore, the recovery of Li from

the spent batteries has become increasingly important from both economic and environmental point of views.

Several methods to recover the metal from spent batteries include metal leaching (Sun and Qiu, 2012; Meshram et al., 2015), physical separation (Bertuol et al., 2015), mechanochemical (Tan and Li, 2015), and Co and Li separation (Joulié et al., 2014). Hydrometallurgy is commonly used to extract metals from spent batteries (Chen et al., 2015). During the early stage of recovery, battery electrodes are leached in a strong acid solution, such as H₂SO₄ (Meshram et al., 2015), HCL (Guo et al., 2016), succinic (Li et al., 2015), oxalic (Zeng et al., 2015) and tartaric acids (Nayaka et al., 2016), to obtain a mixture of metal ions mostly containing Li, Co, Ni, Mn, and several others, depending on the battery type. Unfortunately, although these acids lead to relatively good results, they are corrosive, environmentally polluting, generating liquor waste, and causing difficult post treatment (Wang et al., 2016; Pant and Dolker, 2017). In addition, after diluting the metals, the ion species must be separated and purified by available technologies such as multistage precipitation, electrodialysis (ED) and selective adsorption (Tuncuk et al., 2012).

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Nomenclature

C_{out}	column outlet concentration	$t_{0.5}$	breakthrough time
C_{in}	initial concentration	$t_{0.5}^2$	breakthrough time of the second ion to appear
t	adsorption time	t_{lag}	lag time

The conventional precipitation method requires chemicals to properly adjust the solubility of each component. The method consists of several steps of mixing, heating and filtration and involves a careful stepwise increase of the pH by the addition of a basic solution (NaOH or Na_2CO_3), followed by evaporation and filtration at every step to precipitate the specific metal ion. Meanwhile, an ED requires membrane and electric current to separate the ion mixtures. Thus, the technology is still expensive and prone to membrane fouling. On the other hand, metal adsorption using activated carbon (AC) from biomass waste is considered to be an effective method, especially in acidic environment (Chand et al., 2009).

The recovery of Li ions using various adsorbents has been investigated mainly for extracting Li from brine (Ooi et al., 2016; Xiao et al., 2015). The focus of most studies is to produce a novel adsorbent whose uptake capacity of Li ions is as high as possible. Some of the previous efforts have been tested using pure Li solution to observe the performance of the adsorbents (Jeong et al., 2015; Zhang et al., 2016). Li is a light element that is quite difficult to be physically adsorbed, compared to other possible metal ions present in brine or spent Li-ion battery leachate solutions. Commonly, the interaction between Li ion and the adsorbent surface is the weakest among the other present metal ions. To enhance the uptake of Li ion from solution, some advanced materials have been prepared as adsorbents (Xiao et al., 2015; Lemaire et al., 2012). However, it should be kept in mind that the proposed Li adsorbents have only been tested in single-ion solutions. Therefore, these methods may have the drawback of stronger affinity towards other metals when applied in a multicomponent metal ion solution. If this is the case, the separation effect of Li from other metals will not be attained.

In this study, to answer the above-mentioned problems, a novel column adsorption method to separate Li from other dissolved metal ions was developed. The success of the separation depends on the adsorbent, whose adsorption affinity for Li should be greatly different from that for other metal ions. The adsorption experiments were carried out using a synthetic leaching solution containing Li, Mn, Ni, and Co ions. In addition, a Li-ion battery electrode leaching solution was also used after understanding the sorption behavior of the adsorbent in the preliminary batch tests.

In the present work, an AC from the low-cost precursor of coconut shell charcoal was selected as adsorbent. AC is considered as an effective adsorbent mainly due to its large surface area and good adsorption capability. Unfortunately, high production cost generally becomes barrier in its adoption; therefore, a low cost AC is extremely encouraged. AC from coconut shell charcoal has been adopted for several separation/adsorption processes, including Ni (Jeong et al., 2015), phenol (Karri et al., 2017), benzene, toluene (Mohammed et al., 2015), sulfamethoxazole (Tonucci et al., 2015), and Pb (Kaccin et al., 2015). However, to the best of authors' knowledge, there is no study dealing with the effort to utilize this kind of AC for Li recovery. In this study, the coconut shell charcoal was activated to enhance its sorption affinity for heavy metal ions (Co, Mn and Ni), while the affinity remained low for Li ions. Such modification was achieved by varying the activation method and temperature to obtain the unique properties of AC. The column adsorption method using the prepared AC aimed at producing a Li-rich solution separated from the leachate mixture of a spent

Li-ion battery, which then underwent precipitation to produce high purity Li_2CO_3 powder. Meanwhile, the other metal ions remained on the adsorbent for further recovery or desorption processes. Thus, the objective of this study is to develop an efficient way for lithium ion recovery by an innovative adsorption column method instead of focusing on the expensive selective adsorbent preparation.

2. Experiments

2.1. AC preparation

Coconut shell char flakes collected from a local charcoal factory were crushed and then sieved to obtain 6–8 mesh (large) and 9–14 mesh (small) fractions, which were stored separately. Furthermore, a portion of the chars was activated by the CO_2 physical activation and KOH chemical activation method. The detail of activation procedures follows a previous report by Purnomo et al. (2012). Typically for physical activation, the process used a temperature variation from 700 to 800 °C in a programmable horizontal tube furnace with increase rate of 20 °C/min under CO_2 gas with a flow-rate of 2 ml/min. A 35 cm long alumina tube with an external diameter of 2.5 cm was used as the char holder during the activation. The char was packed 10 cm long in the middle of the tube with the aid of ceramic wool. The activated char was then washed with deionized water to remove small debris created during the preparation stages that may interfere with the adsorption result, dried in an oven and eventually stored in a sealed container.

Another portion of the prepared coconut shell charcoal was chemically activated using 10 wt% KOH. The char that was treated with the KOH solution was then dried and activated in the same horizontal tube furnace at two temperatures of 700 and 750 °C. Nitrogen was flown through the tube at 2 ml/min during temperature ramp. Once the maximum temperature was reached, the atmosphere was changed to CO_2 (2 ml/min), and the temperature was kept constant for 1 h. After cooling to ambient temperature, the ACs were washed and filtered with deionized water until the filtrate reached a neutral pH. The carbons were then dried in an oven and stored. The samples with different activation methods are coded as listed in Table 1.

2.2. AC characterization

The porosity of the prepared carbon samples was analyzed using a Surface Area Analyzer (NOVA 2000, Quantachrome). The surface morphology of char samples before and after activation was observed by SEM (JSM 6510LA, JEOL). For investigating the

Table 1
ACs sample identification.

ID	Activation method	Activation temp. (°C)
CC	NA	NA
AC700	Physical	700
AC750	Physical	750
AC800	Physical	800
AC700OH	Chemical	700
AC750OH	Chemical	750

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