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Electrochemical process for electrode material of spent lithium ion batteries

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ABSTRACT

Electrochemical method for recovering cobalt and manganese from electrode materials of spent lithium ion batteries was studied. Electrochemical leaching of cobalt and manganese from electrode material was optimized by varying different process parameters such as time, acid concentration and current density. Both cobalt and manganese could effectively be leached out at a current density of 400 A/m² in 3 h using 2 M sulphuric acid. In the subsequent study, the metallic cobalt and electrolytic manganese dioxides was recovered from the leach liquor at 200 A/m², pH 2–2.5 and 90 °C after removing aluminum. The commercial feasibility of the study was tested in pilot scale. Overall recovery of Co, Cu and Mn was above 96%, 97% and 99%, respectively.

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1. Introduction

Lithium-ion batteries (LIBs) play a major role in consumer electronics such as mobile phones, laptops and video cameras due to their high energy density, high power density and long service life (Barik et al., 2016). In addition, demand for LIBs in the automobile industry is also expected to increase with the growing demand for electric vehicles. It is estimated that the global LIBs market is projected to grow at a compound annual growth rate of over 17% during 2016–2021 (Mathews, 2016). Rapid consumption rate of LIBs is an indication for environmental hazards at the end of life unless it is recycled properly (Hunt, 2015). Recycling spent LIBs is beneficial not only for metal recovery but also protecting the environment and conserve primary resources. Moreover, the spent LIBs are regarded as one of the best potential sources of metals such as Co, Mn, Ni, Cu and Al, as their metal values are nearly equivalent to their corresponding primary resources (Barik et al., 2017). Hence, based on the economic value and environmental threat, recycling of spent LIBs is essential and worthy too.

For recycling of spent LIBs several studies have been reported yet (Xu et al., 2008; Wang et al., 2009; Zhang et al., 2013; Barik et al., 2017; Ordonez et al., 2016). Based on the studies reported, the main focus is on the electrode material as it contains the valuable Co, Mn, Cu and Li, and which needs a sustainable recycling

process for their recoveries (Zhang et al., 2013). In these processes, the electrode material was peeled off from the aluminum substrate and leached with various acids followed by separation. In the chemical leaching process of electrode material, acid is required for the dissolution of the metals from the substrate and alkali for separation of valuable metals either by precipitation or solvent extraction (Barik et al., 2017; Hossain et al., 2011). Though solvent extraction process is an industrially preferred process for the separation, it has few drawbacks such as consumption of diluents, solvents and alkalis, risk of fire accident due to low flash point of diluents, environmental issues due to volatile nature of diluents and spillage of solvents. Further, there may be formation of crud in the system while processing secondaries, which will be a huge inventory loss and economically unaffordable for the industries. On the other hand separation by precipitation method (Xu et al., 2008; Ordonez et al., 2016; Yang et al., 2017), though not preferred industrially, it is simple by operation but economically not affordable due to loss of valuable metals by co-precipitation with impurities. Thus there is a need for a process which can overcome the above drawbacks.

Electrochemical processes are simple, easy to control and scale up, clean and environmentally friendly (Prabaharan and Barik, 2016; Paunović and Jordanov, 2011). By considering the standard reduction potentials (Bard et al., 1985; Milazzo et al., 1978) of Co, Cu, Mn and Li, leaching and separation are feasible using an electrochemical method and it has following advantages over other conventional process.

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- (i) Cu can be deposited selectively on cathode due to its positive potential ($E^0 = +0.337\text{ V}$) without the loss of other metal ions (Co, Mn and Li) and does not require any additional chemical for removal/recovery (Gallios and Matis, 1998).
- (ii) Co ($E^0 = -0.28\text{ V}$) and Mn ($E^0 = -1.18\text{ V}$) also can be separated electrolytically due to huge difference in reduction potentials, where as Li ($E^0 = -3.045\text{ V}$) will not deposit electrolytically from aqueous solution due to its negative potential.

Thus the application of electrochemical processes for recovering metal values from electrode material of spent LIBs is worthy to attempt. In order to optimize the leaching conditions, different process parameters such as leaching time, sulphuric acid concentration and current density have been studied. Electrolyte pH, the only critical parameter for electrowinning of Co and Mn was also investigated and discussed in details. Further, the pilot scale study and commercial feasibility of the process were also included.

2. Experimental

2.1. Materials

Spent LIBs (20 kg) having different mA h values were collected from the dismantling section of the Attero Recycling Pvt. Ltd., India. Spent LIBs are shredded in the presence of water because water acts as a scrubbing agent as well as a fire-retardant. After shredding, the output slurry containing plastic/Teflon matrix floats on water and is removed manually, while the other components such as electrode materials, copper foils, aluminum casings/foils, PCBs are separated through a sieve (30 mesh). Except electrode material other components remained over the sieve. The electrode material slurry was filtered and the cake obtained was dried. The dried material was roasted at $500\text{ }^\circ\text{C}$ for 1 h and then taken for the study. Weighed amount of the roasted material was subjected to complete digestion using aqua regia followed by dilution and analysis. Table 1 presents the composition of the sample used in this study. Commercial grade of sulphuric acid (98%), sodium hydroxide and soda ash were used for the study.

Table 1
Chemical position of electrode material of spent LIBs.

Element	Co	Mn	Li	Al	Ni	Cu
%	21.4	10.5	3.74	3.2	0.23	1.3

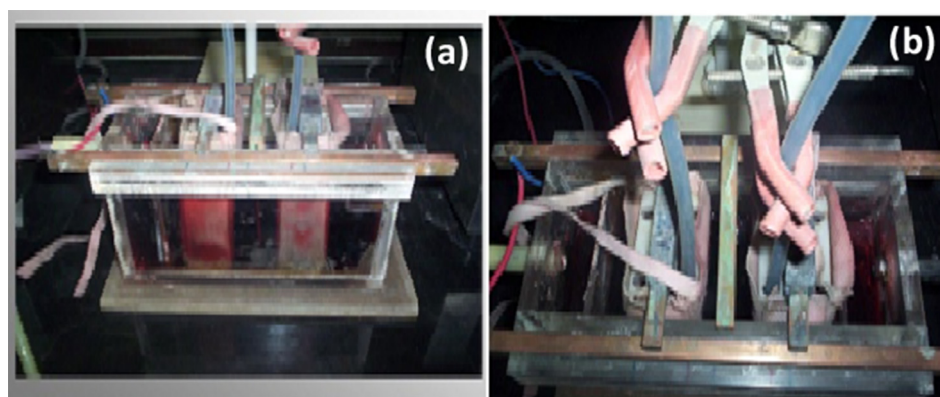


Fig. 1. Side view (a) and top view (b) of the electrolytic cell for leaching of electrode material using anode bags.

2.2. Methods

2.2.1. Electrochemical leaching of cathode materials

Electrochemical leaching experiments (Fig. 1) were carried out in a rectangular cell (capacity 2 L) made of fiber reinforced plastic (FRP) containing anodes (lead, $100 \times 70\text{ mm}^2$) kept inside bags (poly propylene, 100 mesh) and cathodes (SS-316, $110 \times 50\text{ mm}^2$). The electrodes are connected to a rectifier through a copper wire. For each run, 150 g of the sample was added into the bags containing anodes and acidic solution was fed from a 5 L container into the cell through a peristaltic pump and the over flow was collected as leach liquor. Most of the experiments were carried out in duplicate, and the results generally agreed within $\pm 3\%$.

2.2.2. Electrowinning of Co and electrolytic manganese dioxide (EMD)

Electrowinning experiments were carried out in another cell of the similar configuration containing the same numbers of anodes and cathodes. The cell (capacity 2 L) consisted of an inlet for pH adjusted leach liquor/regenerated electrolyte feed and an outlet from where the spent acidic electrolyte overflowed into the regeneration tank. The temperature of the electrolyte was maintained by using an immersion heater (silica cased). The anodes were placed at a constant distance of 50mm from cathodes. A DC power supply was used for passing current in the cell. A peristaltic pump was used to feed the electrolyte in the cell and for maintaining constant flow rate (10 ml/min).

2.2.3. Analytical techniques

The composition of electrode material of spent LIBs and the yield/purity of the product were analyzed with Microwave Plasma-Atomic Emission Spectroscopy (4100 MP-AES), an Agilent instrument except manganese, which was estimated by titration method with EDTA and Eriochrome black-T indicator. A multi-element standard (MP-AES grade) was used for the analysis of the metal ion concentration. X-ray diffraction pattern of the product obtained during the process was investigated using a powder diffractometer (Bruker, D8 Advance) in the range of $10\text{--}80^\circ$ (2θ) at a scanning rate of $2^\circ/\text{min}$. The pH values of the aqueous solutions were measured with a pH/mV meter (Model CL 54+, TOSHCON Industries Pvt. Ltd., and India).

3. Results and discussion

3.1. Electrochemical leaching of cathode materials

Generally, the electrochemical process mainly involves anodic and cathodic reaction. It is well known (Forsén and Aromaa,

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