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Development of a metal recovery process from Li-ion battery wastes

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Abstract

A process for the recovery of lithium and cobalt from the waste of lithium ion batteries using sulfuric acid and hydrogen peroxide was proposed, and metal leaching performance was investigated. The proposed procedure consisted of mechanical separation of metal-containing particles and a chemical leaching process. The effects of leaching agent, of the size of metal-bearing particles, and of incineration as a pretreatment for the leaching, were examined here.

Two stages of crushing and sieving resulted in satisfactory separation of the metal-bearing particles from the waste. Sulfuric acid leaching with hydrogen peroxide in a concentration of 15 vol.% gave a full recovery of the metals within 10 min of processing time. The leaching was carried out at a temperature of 75 °C and with an agitation of 300 rpm for a pulp concentration of 50 g/L in a batch extractor. It was also found that incineration of lithium cobalt oxide particles to remove carbon and organic binder before chemical leaching significantly reduces the leaching efficiency.

1. Introduction

Demand for secondary batteries has steadily increased in recent times as portable electronic appliances such as cellular phones and laptop computers have come into wide use. Along with the development of such portable devices, the technology of secondary batteries–their power source–has rapidly improved in tandem with their applications. In consequence, the amount of secondary battery waste is increasing along with the proliferation of the portable devices. With the disposal of the spent batteries, mostly by landfill, soil contamination follows from the leakage of organic electrolyte as well as heavy metals such as copper and nickel contained in the batteries.

Lithium ion batteries consist of heavy metals, organic chemicals and plastics in the proportion of 5–20% cobalt, 5–10% nickel, 5–7% lithium, 15% organic chemicals and 7% plastics, the composition varying slightly with different manufacturers. When

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the lithium battery wastes are properly processed, valuable metals such as cobalt and lithium can be recovered. From the viewpoints of environmental preservation and recovery of valuable resources, the recycling of spent lithium ion batteries is highly desirable. The current status of the recycling process has been reviewed in several studies (Bernardes et al., 2003, 2004; Espinosa et al., 2004).

Unlike other batteries, lithium ion batteries often blow up during the recycling process due to radical oxidation when lithium metal produced from battery overcharge sustains a mechanical shock from exposure to the air (Contestabile et al., 1999). Therefore, a preliminary separation before recovery of the valuable metals from the waste is necessary. The process also reduces scrap volume, separates battery components and enriches valuable metals. On the other hand, lithium cobalt oxide ($LiCoO_2$)–an active material used as a cathode–is not dissolved easily with common leaching chemicals. Zhang et al. (1998) performed a study of leaching the lithium cobalt oxide with various leachants, and of the effect of leaching condition such as leachant concentration, temperature, leaching time and solid-to-liquid ratio, in hydrometallurgical recoveries of lithium and cobalt. In that study, hydrochloric acid gave the best performance among the three leachants examined, and it has been utilized in other leaching processes for lithium and cobalt (Contestabile et al., 2001). Nitric acid has also been employed instead of hydrochloric acid in other lithium leaching processes (Catillo et al., 2002). A commercial process was introduced and developed by AEA Technology Batteries, though the leachant in which metal lithium and Co(II) ion were electrochemically generated was not specified (Lain, 2001). A new procedure for nitric acid leaching of lithium and cobalt from lithium ion batteries was proposed by Lee and Rhee (2003), and the procedure was extended to regenerate lithium cobalt oxide electrode from the leachate and to examine the performance of the electrode (Lee and Rhee, 2002). Recently, a procedure for the direct regeneration of lithium cobalt oxide electrode from waste lithium ion batteries has been published by Kim et al. (2004).

In this study, a sulfuric acid leaching process for the recovery of lithium and cobalt from lithium ion battery wastes was proposed and its performance was evaluated. The effects of leaching agent, particle size and incineration on acid leaching were investigated to find the optimum condition for metal recovery. The procedure includes a thoroughgoing process of mechanical separation of lithium cobalt oxide particles, which can be applicable in commercial recycling of lithium ion battery waste. Though the hydrometallurgical process of battery recycling has been well developed, preprocessing by mechanical separation has not been so deeply studied as to facilitate the scale-up of the laboratory process for commercialization.

2. Experimental

2.1. Chemicals

In this experiment two reagent grade chemicals were used. Sulfuric acid (Junsei Chemical Co., Japan, Code No. 83010-1230) was utilized for leach-

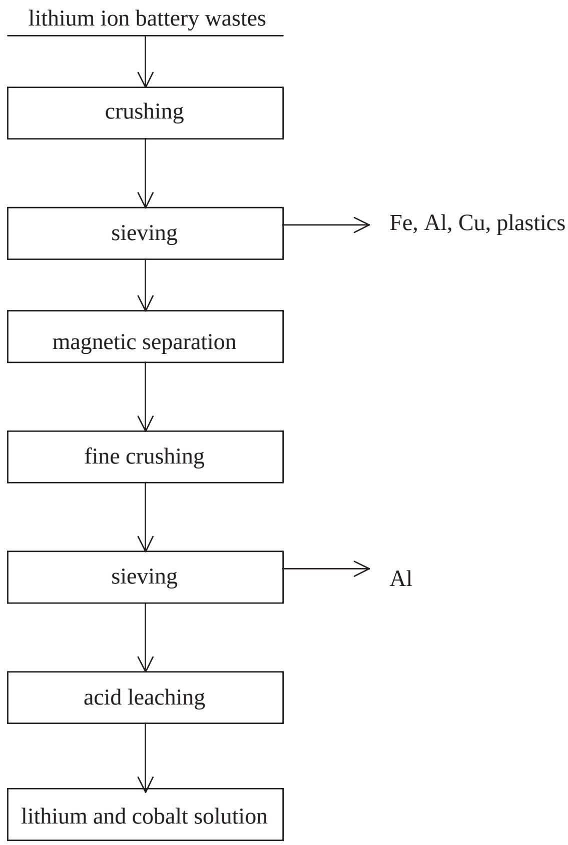

Fig. 1. A flowchart of the proposed metal recovery process.