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Downstream recovery of Li and value-added metals (Ni, Co, and Mn) from leach liquor of spent lithium-ion batteries using a membrane-integrated hybrid system

Ramesh Kumar [a], Chengjia Liu [a], Geon-Soo Ha [a], Young-Kwon Park [b], Moonis Ali Khan [c], Min Jang [d], Sang-Hyoun Kim [e], Mohammed A. Amin [f], Amel Gacem [g], Byong-Hun Jeon [a,*]

[a] Department of Earth Resources & Environmental Engineering, Hanyang University, 222-Wangsimni-ro, Seongdong-gu, Seoul 04763, Republic of Korea
[b] School of Environmental Engineering, University of Seoul, Seoul 02504, Republic of Korea
[c] Chemistry Department, College of Science, King Saud University, Riyadh 11451, Saudi Arabia
[d] Department of Environmental Engineering, Kwangwoon University, Seoul 01897, Republic of Korea
[e] School of Civil and Environmental Engineering, Yonsei University, Seoul 03722, Republic of Korea
[f] Department of Chemistry, College of Science, Taif University, P.O. Box 11099, Taif 21944, Saudi Arabia
[g] Department of Physics, Faculty of Sciences, University 20 Août 1955, Skikda, Algeria

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ABSTRACT

The end-of-life management of a large number of discarded lithium-ion batteries (LiBs) has become a global issue because of the steady increase in their usage every year. In this study, a novel membrane-integrated hybrid system was developed for recycling commercially valuable metals while treating acidic leach liquor obtained from spent LiBs (mostly $LiFePO_4$ type). Alkaline (NaOH and NH_4OH)-pretreated leach liquor (pH adjusted to 5.53) was ultrafiltered to ensure the effective removal of Fe and Al and reduce the turbidity (~1.6 NTU) prior to processing in a nanofiltration system. The nanofiltration membrane (VNF2) was applied successfully to obtain the rejection values of 92.5% (Ni^{2+}), 94.6% (Co^{2+}), and 95.8% (Mn^{2+}) while permeating > 89.6% of Li^+ with 7.5 L/m^2·h of flux under optimized conditions of transmembrane pressure (10 bar) and crossflow rate (2.25 m^3/h) in the recirculation mode. The fractionation of monovalent ions from bivalent ions was performed in a concentrated mode to enrich the bivalent metal ions, Ni^{2+}, Co^{2+}, and Mn^{2+} from 0.74, 0.52, and 0.63 g/L to 6.14, 4.59, 5.62 g/L, respectively, at 90% recovery of the feed solution. The Li^+ (21.1 g/L) that was contained in the nanofiltrate permeate stream was crystallized into Li_2CO_3; a purity of 99.5 wt% was obtained at 88.2% recovery using 4 M K_2CO_3 at an operating temperature of 70 °C. Hence, the proposed novel system can lead to the development of a clean and sustainable process for the recycling of precious metal ions from end-of-life LiBs for reuse on a commercial scale.

1. Introduction

Lithium-ion batteries (LiBs) have been identified as being crucial to making a fossil fuel-free economy, which constitutes ~ 37% of the world market for rechargeable batteries. This percentage use is due to the beneficial properties of lithium, including a high redox potential (93.045 V) and specific heat capacity (3.489 J/g mol at 20 °C) [1,2] The demand for LiBs for their use in sophisticated electronics, wireless communication devices, portable instruments, cellular phones, laptops, modern electric and hybrid vehicles, and medical equipment has been growing daily. The demand may be attributed the high energy density storage, long cycle life, low self-discharge, stable flow of electricity, and eco-friendly powering of the LiBs [3,4]. Thus, rechargeable LiBs is a promising clean technology that could replace traditional fossil fuel powered devices, especially in the transport sector, to reduce the emission of greenhouse gases in compliance with the Paris Agreement-2015 [5]. LiBs may be fabricated using various conducting metals, *viz.* Li, Co, Ni, Mn, Cu, Fe, and/or Al in different concentrations within compounds including, Li-Co-oxide, Li-Mn-oxide, Li-Ni-Mn-Co-oxide, Li-Ni-Co-Al-oxide, and Li-Fe-phosphate [6,7].

Li^+, along with other key metal ions (Ni^{2+}, Co^{2+}, and Mn^{2+}) associated with LiBs is of significance because of the limited availability of

* Corresponding author.
E-mail address: bhjeon@hanyang.ac.kr (B.-H. Jeon).

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