INTERNATIONAL JOURNAL OF
Minerals, Metallurgy and Materials

Volume 32, Number 12, December 2025
Page 2958

Research Article https://doi.org/10.1007/s12613-025-3216-5

Chelating extraction of critical metals from cathode of end-of-life lithium titanium oxide batteries: Experiments, machine learning and validation

Heewon Kang[1),*], *Muhammad Farhan*[1),*], *Sohwi Park*[1)], *Li Cai*[2)], *Allan Gomez-Flores*[1),✉], and *Hyunjung Kim*[1),✉]

1) Department of Earth Resources and Environmental Engineering, Hanyang University, 222 Wangsimni-ro, Seongdong-gu, Seoul 04763, Republic of Korea
2) College of Environmental Science and Engineering, Donghua University, Shanghai 201620, China
(Received: 27 April 2025; revised: 12 June 2025; accepted: 24 June 2025)

Abstract: Lithium-ion batteries (LIBs) that reached their end-of-life (EoL) require recycling, rather than disposal, to recirculate valuable metals and protect the environment. This led us to investigate the extraction of metals from the cathodes of EoL lithium-titanate batteries using ethylenediaminetetraacetic acid disodium (EDTA-2Na). In this work, an orthogonal array was used to design experiments and signal-to-noise calculations were used to define the optimal conditions, which were 0.50 mol/L EDTA-2Na, pH = 6, 75°C, 180 min, 2% pulp density, and 300 r/min, resulting in 97.96%, 94.79%, 96.45%, and 98.89% leaching efficiencies for Li, Ni, Co, and Mn, respectively. Statistically significant interactions between variables were then identified using Pearson's correlation at the 95% confidence interval, and the pH and temperature were found to be significant. The extraction efficiency decreased as the pH increased, but increased as the temperature increased. Machine learning fitting using linear regression for multi-output prediction was unsatisfactory, whereas random forest regression (RFR) produced satisfactory results. Permutation importance was computed on the fitted RFR to determine feature importance, and confirmed that the pH and temperature were influential variables; however, the time and pulp density were also noted. As the fitted RFR failed to satisfactorily predict leaching efficiencies in additional validation experiments, we recommend increasing the number of experiments and using additional fitting models. An additional analysis that included the initial oxidation–reduction potential (optimal 33.3 mV) revealed this to be the most important variable, the effect of which largely overshadows those of all the other variables. Finally, an environmental assessment highlighted the benefits of the chelating extraction; however, the economic assessment indicated room for improvement.

Keywords: end-of-life lithium-ion battery; lithium titanium oxide battery; hydrometallurgy; chelating extraction; machine learning

1. Introduction

Technological development, the depletion of nonrenewable resources, and pollution have increased the demand for green energy, particularly in the form of lithium-ion batteries (LIBs) to power electric vehicles (EVs) [1–2]. The production of LIBs experienced remarkable growth in the decades following the 1990s because of their high specific energy (>250 W·h/kg), high specific power (>1000 W/kg), long life span (up to ~10000 cycles), and wide operating temperature when discharging (20–55°C) [3–4]. Globally, a 25% increase in EV sales in 2024 versus 2023, along with a ~12% growth in the production of LIBs, may be related to the increased demand for Li, Ni, and Co by 5.9%, 1.5%, and 3.1%, respectively, as projected for the period from 2021 to 2040 [5–7]. However, the growing uptake of LIBs poses a major environmental burden because of their disposal once their end-of-life (EoL) is reached [1–2,8]. Spent LIBs contain 5wt%–7wt%, 5wt%–10wt%, and 5wt%–20wt% of Li, Ni, and Co, respectively, which exceed the concentrations of these metals in natural ores and makes recycling economically attractive [9–10]. Moreover, the global market for recycling spent LIBs is estimated to rise from 16.2 billion USD in 2024 to 56.9 billion USD by 2032 [11].

Recycling spent LIBs is crucial to protect the environment because of the toxic electrolytes and heavy metals (Ni, Co, and Mn) these batteries contain, and to meet the demand by reusing these valuable metals [12–13]. Hydrometallurgy, pyrometallurgy, froth flotation, and direct regeneration are methods that are used to recycle the anode and cathode active materials of LIBs [14–16]. Pyrometallurgy uses high temperatures to recover metals with a yield of >90% through the formation of Ni, Co, and Cu alloys but fails to effectively recover Li, graphite, and the electrolytes [17]. Flotation involves the grinding and pyrolysis of electrode active materials from spent LIBs at mild temperatures. Bubbles in agitated tanks are then used for the selective separation of graphite and Li oxides based on their hydrophobicity differences,

*These authors contributed equally to this work.
✉ Corresponding authors: Allan Gomez-Flores E-mail: allanchito@gmail.com; Hyunjung Kim E-mail: kshjkim@hanyang.ac.kr
© University of Science and Technology Beijing 2025
Springer