



Hydrometallurgical recycling of lithium from waste saggar: Studies on influential role of acid/alkaline additives, leaching kinetics and mechanism

Wonhong Song, Sowon Choi, Hyunjung Kim^{*}, Sadia Ilyas^{*}

Department of Earth Resources & Environmental Engineering, Hanyang University, Seongdong-gu, Seoul 04763, the Republic of Korea

ARTICLE INFO

Editor: Dr. G. Palmisano

Keywords:

Energy-critical element
Lithium
Additive leaching
Waste saggar
Resource recycling

ABSTRACT

Waste recycling has become crucial with the increasing attention paid to environmental rules and the fast-depleting resources of critical elements. Soaring demands for lithium in cathode materials of rechargeable batteries result in a surge in waste saggar after being used in the calcination of cathode precursor. The recycling of lithium from waste saggar is therefore imperative due to the high supply-risk of lithium. Herein, waste saggar composed of various mineral phases (26.26 % $\text{LiAlSi}_4\text{O}_{10} \cong 0.6\%$ Li; 6.34 % $\text{Li}_4\text{SiO}_4 \cong 1.47\%$ Li; 0.79 % $\text{LiOH} \cdot \text{H}_2\text{O} \cong 0.13\%$ Li; 0.03 % $\text{Li}_2\text{CO}_3 \cong 0.01\%$ Li) was leached in water for lithium extraction; while the effects of NaOH, $\text{Ca}(\text{OH})_2$, and H_2SO_4 as potential additives were examined under a wide temperature range of 5–80 °C. Interestingly, the observed order of leaching efficiency at lower temperature (20 °C) $\text{H}_2\text{SO}_4 > \text{NaOH} > \text{Ca}(\text{OH})_2$ was in variance with that at 80 °C ($\text{NaOH} > \text{H}_2\text{SO}_4 > \text{Ca}(\text{OH})_2$). The apparent activation energy for lithium extraction was determined to be 29.8 kJ/mol using NaOH, 33.8 kJ/mol using $\text{Ca}(\text{OH})_2$, and 14.1 kJ/mol using H_2SO_4 , indicating that the overall leaching process follows a mixed-controlled mechanism. The maximum efficiency of 94 % Li-extraction was achieved with NaOH which was used for the precipitation recovery of Li_2CO_3 of purity above 99 %.

1. Introduction

Lithium-ion batteries (LIBs) offer advantageous properties with high energy density, long durability, lightweight, and small volume over other rechargeable batteries of commercial usage [10,11,19,3]. Since the advent of LIBs, a paradigm shift in energy generation and storage technology has been observed, particularly in portable electronic devices and electric vehicles (EVs) [21,41]. The demands for LIBs have grown rapidly because of the frequent change in consumer electronics engineering and the global shifting towards EVs [17,23]. The annual sales of EVs in 2030 are projected to be 21–31 million [30]. Consequently, the soaring demands for LIBs are expected to increase by 14-folds between 2020 and 2030 [7]. Consequently, it would result in high lithium demand and a supply crunch in the commodity market.

Securing lithium from alternative sources is in high traction for metallurgical researchers. For example, its extraction from geothermal water [33] and seawater [34], albeit these techniques still face economic, environmental, and scale-up challenges. Recycling of spent LIBs has been widely explored as a secondary resource to establish the circular economy of lithium and other critical metals therein [2,13]. This

source depends on the generation of post-consumer waste volume, which is always accompanied by an increased production of battery materials [24,26]. The preparation of cathode materials into saggar pot is a significant step in this process; thereby, the precursors are subjected to the calcination at a higher temperature forming the product viz., $\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$ (LNCM), LiCoO_2 (LCO), and $\text{Li}_m\text{Ni}_x\text{Co}_y\text{Al}_z\text{O}_2$ (LNCA). As the saggars are made of ceramic materials, the inner wall comes in direct contact with the precursor compounds/elements during the progress of the chemical reactions [22,27,36]. It also drives the refractory material forming new multi-element compounds on the saggar-wall surface [39]. Because of the usual excess mixing of LiOH with transition metal oxides, the solid-state reaction induced at a high temperature, in which molten lithium component penetrates and spreads into the inner wall of saggar, prominently forms the composite oxides, Li–Al–Si–O [14]. In this manner, the repeated usage of saggar in cathode production promotes surface corrosion and decreases efficacy with an increased impurity profile. Consequently, the replacement of saggar at regular intervals becomes a necessity for a higher conversion and efficient yielding of the cathode mass. Commonly, after each 5–20 cycles, the used saggar box has to be scrapped as a waste [40].

^{*} Corresponding authors.

E-mail addresses: kshjkim@hanyang.ac.kr (H. Kim), sadiailyas1@yahoo.com (S. Ilyas).

<https://doi.org/10.1016/j.jece.2023.110407>

Received 15 March 2023; Received in revised form 23 May 2023; Accepted 20 June 2023

Available online 21 June 2023

2213-3437/© 2023 Elsevier Ltd. All rights reserved.

Hydrometallurgical recycling of lithium from waste saggar: Studies on influential role of acid/alkaline additives, leaching kinetics and mechanism

Wonhong Song, Sowon Choi, Hyunjung Kim  , Sadia Ilyas  

Show more 

 Add to Mendeley  Share  Cite

<https://doi.org/10.1016/j.jece.2023.110407> 

[Get rights and content](#) 

Highlights

- Reclamation of Li from waste saggar is proposed using water and additive leaching.
- Leaching order $\text{H}_2\text{SO}_4 > \text{NaOH} > \text{Ca}(\text{OH})_2$ at 20°C altered to $\text{NaOH} > \text{H}_2\text{SO}_4 > \text{Ca}(\text{OH})_2$ at 80°C .
- E_a values, 29.8–33.8 kJ/mol indicate mixed-controlled reaction for additive leaching.
- Maximum leaching efficacy of 94 % with NaOH additive yielding 3.9 g/L Li in solution.
- Li_2CO_3 of more than 99 % purity is precipitated from the leach liquor using Na_2CO_3 .