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Recovering Li from the acid leachate of spent Li-ion batteries by hydrometallurgical separation and improving the performance by nano-filter pre-treatment

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Lithium (Li) is usually obtained mainly from brine, ores, and seawater [1]; however, to reduce the environmental burden caused by resource development, it is necessary to establish Li recycling from spent Li-containing products. Li-ion batteries (LiBs) have a high Li content, and the supply is rapidly increasing worldwide. Thus, spent LiBs have a high potential as a secondary Li resource [1]. In the recycling process, spent LiBs are crushed and separated, then become a powder containing various metals, called black mass [2]. After dissolving it with acid, various metals are precipitated sequentially and recovered [2]. The traditional sequential precipitation method has the problem of a decrease in the recovery rate of Li by co-precipitate. Thus, we attempted to achieve an effective Li recovery process by pre-separating Li from the acid leachate of LIB using a novel nano-filter with acid tolerance produced by TORAY Industries, Inc. (Tokyo, Japan). In this study, we evaluate the effect of the nano-filter from the precipitation behavior of each ion in the separation process, and the factors of Li-loss are clarified.

This study used three types of acid leachate from LiBs black mass, provided by different recyclers and their respective nano-filter permeates. Firstly, $Ca(OH)_2$ was added to eliminate sulfate and multivalent cations. Na_2CO_3 was then introduced to remove excess Ca^{2+} and supply carbonate ions, and Li was finally recovered by removing the solvent by evaporation.

The total amount of Li was lost as the solution content in the solid phase in the first precipitation step for all types of samples. However, the lost percentages for the nano filter permeate were negligible. Meanwhile, Li was also lost by coprecipitated with CaSO4 and Al minerals. In the carbonation process, 20-30% of Li was lost in the raw sample, whereas the loss rate in the nanofiltration membrane permeates was less than 10%. Nanofiltration membrane treatment reduced the amount of added reagents and suppressed Li loss, indicating that nano-filtration is a quantitatively effective process for Li recovery.

[1] Swain, *Sep. Purif. Technol.*, 2017.
[2] Saleem et al., *Chem. Eng. J. Adv.*, 2024.