Modeling Framework for Optimization of the Life Cycle Sustainability of Lithium-Ion Batteries by Nickel Manganese Cobalt Recycling Process

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Abstract

The increasing utilization of lithium nickel manganese cobalt oxide (NCM) batteries has led to a greater demand for the retrieval of cathode materials to support sustainable battery recycling. This is essential due to the finite availability of metals and the need to reduce the adverse environmental consequences associated with battery disposal. Despite attempts to enhance the sustainability of Lithium-Ion Batteries (LIB) life cycle, few studies have comprehensively addressed both economic and environmental aspects. Several end-of-pipe chemical processes for recycling LIB have been proposed, along with their techno-economic analyses, without considering their environmental sustainability. Thus, this study introduces multi-objective optimization considering economic profit and life cycle environmental performance simultaneously by applying it to optimize the NCM battery recycling process. A hydrometallurgical intensified process model of LIB recycling was developed using a process simulator Aspen Plus, transforming LIB waste into NCM hydroxide. A model of the cradle-to-gate life cycle of LIBs was developed in the LCA software OpenLCA. Following, based on multi-objective Bayesian optimization, the optimal pareto points were identified. The results of this study make decisions on selecting and optimizing battery recycling processes, contributing to achieving both economic viability and sustainability while closing the material loop of LIBs. Also, this is the first study which interconnects Aspen Plus with OpenLCA to perform optimization, thereby contributing to the potential development of automated life cycle assessments.

**Keywords**: Life Cycle Assessment Automation, Lithium-ion Battery Recycling Process, Circular Economy, Techno-economic Analysis, Bayesian Optimization

* 1. Introduction

Concerns regarding resource depletion and environmental impacts of battery waste pose a potential obstacle to a sustainable energy transition (Mohr et al., 2020). Thus, recycling waste batteries is crucial to manage the expected stream of discarded batteries and minimize the potential depletion of critical resources such as lithium, manganese, and cobalt. Hydrometallurgical processes stand out as primary LIB recycling technologies. This process involves the use of acids to dissolve the metals of the cathode and recovers the targeted metals by selective precipitation. Despite its high recovery efficiency and no gaseous pollutants through this process, this has some limitations concerning its environmental footprint due to its substantial space requirement and wastewater production.

Efforts to address these limitations have led to the proposal of novel hydrometallurgical processes and subsequent assessment of their environmental impacts using life cycle assessment methodology to identify environmental hotspots (Arshad et al., 2022; Dewulf et al., 2010; Kim et al., 2022; Kim et al., 2023; Quan et al., 2022). The research from Kim et al. (2023) proposed and simulated an innovative LIB recycling process integrated with a hydrogen roasting and carbon dioxide waste gas stream generated from a combustion process for district heating. This novel process achieves 98.10% Li recovery without emissions and demonstrated economic feasibility, surpassing the net present value of the proposed process by 3.08% compared to conventional hydrometallurgy. In addition, Du et al. (2022) present a life cycle assessment of the Chinese hydrometallurgical process using the ReCiPe 2016 methodology. This reveals that the leaching and extraction process and pretreatment process contributed 35.08% and 34.66% to the Global Warming Potential (GWP) of the process.

In this context, this study proposes an intensified hydrometallurgical process, modelled using Aspen Plus, aiming to maximize economic profits of NCM hydroxide (the end-product of this process) and minimize potential environmental impacts of the process simultaneously. While maximizing the product yield is essential with respect to the economic feasibility and profitability of the targeted process, it is crucial to recognize that this may exacerbate the possible environmental impact of the process. The outcomes of this study can be used to serve as valuable insights for making informed decisions regarding selecting and optimizing battery recycling processes (Guillén-Gosálbez et al., 2019; Köck et al., 2023).

* 1. Method
     1. Process description

Figure 1 illustrates the schematic diagram detailing both the conventional NCM recycling process and the proposed recycling process (Kim et al., 2023). While the entire process comprises multiple unit processes aimed at recuperating metals from spent LIB cathodes, the proposed process utilizes a single batch process that produces NCM hydroxide. This process uses an optimized sequencing batch reactor cycle comprising three main steps: leaching process, crystallization, and filtration. Initially, the spent batteries are subjected to a leaching process using sulfuric acid and hydrogen peroxide chemicals. Following leaching, the pH of the leachate solution containing Cobalt ions, Nickel ions, and Manganese ions is adjusted to pH 10.6 by adding sodium hydroxide and ammonia. These chemicals facilitate co-precipitation to form NCM hydroxide. The resulting ferrous solid mixture is precipitated as a single clump and then filtered. Any remaining components are considered to be dissolved in wastewater.

|  |  |
| --- | --- |
| (a) |  |
| (b) |  |

Figure 1 Schematic Diagram of Conventional Hydrometallurgical NCM Recycling Process (a) and Proposed NCM Recycling Process (b)

The crystallization process following leaching was modeled using the population balance equation (Hu et al., 2004). The population balance equation describes the evolution of population density under nucleation and particle growth.

where represents slurry volume, denotes the population density of crystals, signifies the particle size, stands for the crystal growth rate, is the nucleation rate, and represents the Dirac delta function. This model determines the evolution of crystal size distribution.

The crystal growth rate is a function of temperature, crystal size, and supersaturation ratio.

where , , , , and are parameters to be estimated by experimental data and S denote the supersaturation ratio.

The methodology employed simulates the crystal size distribution evolution in each time step. Hence, the population density of the precursor is as follows.

and and represent particle size points and slurry volume at time interval .

When calculating the economic profit of this process, only operating expenses were considered, given that this process is based on a single unit batch reactor. The economic profit was determined as the difference between the market price of the NCM products and the total sum of input streams price.

* + 1. Life cycle assessment

The environmental assessment was performed according to the ISO 14040/44. The goal of this study is to assess the environmental impact of the proposed hydrometallurgical recycling process. This assessment follows a cradle-to-gate system boundary. The functional unit is defined as 1 g of NCM hydroxide produced. The life cycle was modelled using OpenLCA software developed by Greendelta, Germany.

The life cycle inventory of the foreground system was retrieved from stream results and operating conditions from the proposed process using pywin32 Python library. This retrieved inventory was processed and loaded onto OpenLCA software via olca-schema and olca-ipc Python libraries. In addition, the LCA database Ecoinvent 3.91 was used to quantify the environmental impacts of the background system. It is assumed that the waste and the wastewater do not undergo the subsequent treatment process. In other words, the waste and the pollutants in the wastewater are considered emissions in this study.

The life cycle impact assessment was done using IPCC 2013 GWP 100a calculation methodology. Global warming was selected as indicator for this study because it is a major public concern. Also, the GWP indicator can be used to help decision-making when selecting one technology over the alternative.

* + 1. Mathematical formulation for optimization

The original design problem can be mathematically formulated as follows.

where and refer to the objective functions (i.e., economic profit and environmental indicator (GWP)), x denotes 15 decision variables (5 scenarios and 3 operating conditions), and LB and UB refer to the lower and upper bounds on the continuous variables, respectively.

Here, the multi-objective Bayesian optimization was used to maximize the economic profits and minimize the environmental impacts using botorch Python library. This is a sequential model-based optimization technique used for optimizing expensive-to-evaluate black-box functions. The surrogate model utilized is the gaussian process regression. Also, the acquisition function used is the expected improvement.

* 1. Results and Discussion

Table 1 and Figure 2 present the outcomes derived from the optimization process, highlighting key findings. Economic profits plateau at 890 US dollars per one batch process operation, while the global warming potential reaches a minimum of 140 kg CO2 eq. The optimization also reveals a peak recovery efficiency of approximately 80% for nickel, cobalt, and manganese, resulting in a maximum total recovery of 500 g NCM hydroxide per 1kg of spent batteries. Figure 2 emphasizes an inverse relationship between economic profit and global warming potential. Notably, the use of NH3 in the process significantly impacts global warming due to its energy-intensive production, emitting substantial carbon dioxide. Reducing NH3 consumption can mitigate these environmental effects.

Conversely, NH3 functions as a chelating agent facilitating the recovery of nickel, cobalt, and manganese, leading to increased NCM hydroxide production and, subsequently, higher economic profits. Therefore, decision-makers need to balance NH3 usage for optimal outcomes. Operational time is another crucial factor. Prolonged operational time increases NCM hydroxide crystallization, augmenting its production and economic benefits. However, this also amplifies environmental impacts, notably the consumption of steam produced by burning coal. The longer operational time results in more coal burning, elevating the global warming potential. Thus, decision-makers must consider both economic and environmental factors when determining operational time, guided by their knowledge and priorities.

Table 1 Process Optimization Result

|  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Optimum | #1 | #2 | #3 | #4 | #5 | #6 | #7 | #8 | #9 |
| Economic Profit (US $) | 846.7 | 851.6 | 856.5 | 857.4 | 857.6 | 858.1 | 875.3 | 880.1 | 890.2 |
| GWP  (kg CO2 Eq.) | 148.0 | 169.2 | 204.6 | 216.7 | 218.4 | 232.6 | 243.3 | 253.7 | 253.7 |

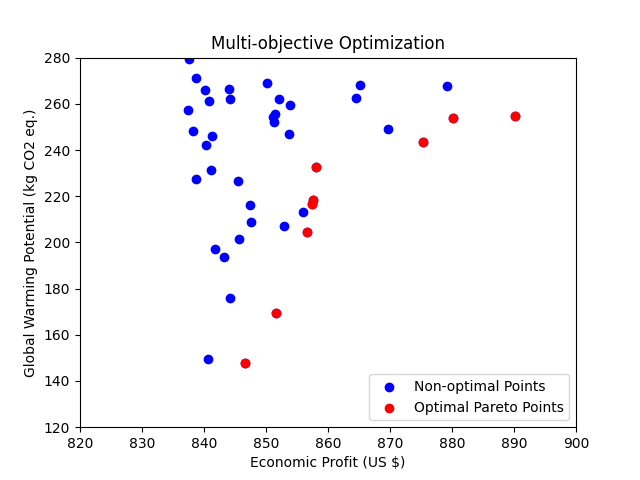


Figure 2: Multi-objective Bayesian Optimization Pareto Points

* 1. Conclusion

This work introduces an intensified hydrometallurgical LIB recycling process with the primary goal of maximizing the output of NCM hydroxide while simultaneously reducing the potential environmental footprint of the process. The NCM hydroxide output is expected to be used in the LIB manufacturing process, replacing some of the resources from mining.

The optimization of this process yields optimal profits ranging from 840 US dollars to 890 US dollars. However, it is noteworthy that as profits increase, the environmental impact, measured by the Global Warming Potential (GWP), also increases, ranging from 148 kg CO2 eq to 254 kg CO2 eq. It is crucial to recognize that while maximizing profitability is a key goal, it comes with the trade-off of heightened environmental consequences. Therefore, maintaining a balance between economic gains and environmental sustainability is essential for achieving long-term operational viability.

This study provides valuable insights that can guide decision-makers in selecting a battery recycling operational strategy. By considering the interplay between economic outcomes and environmental impacts, stakeholders can make well-informed decisions, contributing to the development of sustainable practices in the field of LIB recycling.

References

Arshad, F., Lin, J., Manurkar, N., Fan, E., Ahmad, A., Tariq, M. un N., Wu, F., Chen, R., & Li, L. (2022). Life Cycle Assessment of Lithium-ion Batteries: A Critical Review. In *Resources, Conservation and Recycling* (Vol. 180). Elsevier B.V. https://doi.org/10.1016/j.resconrec.2022.106164

Dewulf, J., Van der Vorst, G., Denturck, K., Van Langenhove, H., Ghyoot, W., Tytgat, J., & Vandeputte, K. (2010). Recycling rechargeable lithium ion batteries: Critical analysis of natural resource savings. *Resources, Conservation and Recycling*, *54*(4), 229–234. https://doi.org/10.1016/j.resconrec.2009.08.004

Du, S., Gao, F., Nie, Z., Liu, Y., Sun, B., & Gong, X. (2022). Life cycle assessment of recycled NiCoMn ternary cathode materials prepared by hydrometallurgical technology for power batteries in China. *Journal of Cleaner Production*, *340*. https://doi.org/10.1016/j.jclepro.2022.130798

Guillén-Gosálbez, G., You, F., Galán-Martín, Á., Pozo, C., & Grossmann, I. E. (2019). Process systems engineering thinking and tools applied to sustainability problems: current landscape and future opportunities. In *Current Opinion in Chemical Engineering* (Vol. 26, pp. 170–179). Elsevier Ltd. https://doi.org/10.1016/j.coche.2019.11.002

Hu, Q., Rohani, S., Wang, D. X., & Jutan, A. (2004). Nonlinear kinetic parameter estimation for batch cooling seeded crystallization. *AIChE Journal*, *50*(8), 1786–1794. https://doi.org/10.1002/aic.10163

Kim, J., Kim, S., Lim, J., Moon, I., & Kim, J. (2022). Sequential flue gas utilization for sustainable leaching and metal precipitation of spent lithium-ion battery cathode material: Process design and techno-economic analysis. *Journal of Cleaner Production*, *380*. https://doi.org/10.1016/j.jclepro.2022.134988

Kim, J., Kim, Y., Moon, I., Cho, H., & Kim, J. (2023). Process design and economic analysis of hydrogen roasting integrated with CCU for a carbon-free spent LIB recycling process. *Chemical Engineering Journal*, *451*. https://doi.org/10.1016/j.cej.2022.139005

Kim, J., Moon, I., & Kim, J. (2023). Integration of wastewater electro-electrodialysis and CO2 capture for sustainable LIB recycling: Process design and economic analyses. *Journal of Cleaner Production*, *391*. https://doi.org/10.1016/j.jclepro.2023.136241

Köck, B., Friedl, A., Serna Loaiza, S., Wukovits, W., & Mihalyi-Schneider, B. (2023). Automation of Life Cycle Assessment—A Critical Review of Developments in the Field of Life Cycle Inventory Analysis. In *Sustainability (Switzerland)* (Vol. 15, Issue 6). MDPI. https://doi.org/10.3390/su15065531

Mohr, M., Peters, J. F., Baumann, M., & Weil, M. (2020). Toward a cell-chemistry specific life cycle assessment of lithium-ion battery recycling processes. *Journal of Industrial Ecology*, *24*(6), 1310–1322. https://doi.org/10.1111/jiec.13021

Quan, J., Zhao, S., Song, D., Wang, T., He, W., & Li, G. (2022). Comparative life cycle assessment of LFP and NCM batteries including the secondary use and different recycling technologies. *Science of the Total Environment*, *819*. https://doi.org/10.1016/j.scitotenv.2022.153105