Designing Li-ion Battery Recycling Networks

Pablo Bernal,a,b Pablo Miranda,c Nathalie Jamett,a Francisco Tapiaa,c, Sebastián Herrera-León,a,b,d\*

aLithium I+D+i Center, Universidad Católica del Norte, Antofagasta 1270709, Chile

bDepartamento de Ingeniería Química y de Medio Ambiente, Universidad Católica del Norte, Antofagasta 1270709, Chile.

cDepartamento de Ingeniería Industrial, Universidad Católica del Norte, Antofagasta 1270709, Chile

dSchool of Engineering Science, LUT University, P.O. Box 20, FI-53851 Lappeenranta, Finland

\*Corresponding author at sebastian.herrera@ucn.cl

Abstract

The transition to electromobility brings a strong demand for lithium-ion batteries that will inevitably result in a large amount of waste in the coming future after these batteries reach their end-of-life. A mixed integer linear program was formulated for designing recycling networks of lithium-ion batteries, comparing the performance of pyrometallurgical and hydrometallurgical recycling processes. The conceptual recycling network proposed consists of three nodes: collection centers, recycling processes, and consumption points. According to the model, the recycling of batteries constitutes 57% of the total cost of the network. In addition, the proposed model only considers the recycling of the cathode material. Therefore, it is crucial to develop recycling networks to process all the battery components efficiently. The proposed model is a valuable tool for designing the supply chain network for recycling Li-ion batteries.

**Keywords**: Recycling network, lithium-ion batteries, MILP

* 1. Introduction

Electromobility is an essential driver in the global development of the automotive industry nowadays since modifying energy sources to mitigate the effects of pollution caused by fossil fuel usage is imperative (Ma et al., 2021). According to (Chakraborty and Saha, 2022), 5.6 million electric vehicles (EVs) circulated globally in 2019, and 58% of the world's vehicle fleet will be EVs by 2040. The growing EV manufacturing requires a significant source of electric batteries, such as Lithium-Ion Batteries (LIBs). Raw materials employed to produce LIB, mainly lithium, cobalt, and graphite, are considered critical or strategic supplies, and therefore, one strategy to dampen their demands and final disposal after LIB usage is to recycle LIBs to recover as much of these materials as possible (Rinne et al., 2021).

A significant challenge when recycling LIB is associated with treating the different chemical components of the cathode material. To achieve high recovery rates, designing distinct processes for each type of battery is required (Mossali et al., 2020). According to this context, this work aims to develop an optimization model for designing a LIB recycling supply chain network. As a first approach, the model compares various recycling methods, considering variables such as recovery efficiency and processing costs.

* 1. Methods

A mixed integer linear programming (MILP) problem is developed to design a recycling network of LIBs. The recycling network comprises three stages: a set of collection facilities, a set of recycling processes, and a set of consumption points. In the first stage, the LIBs are sorted at the collection facilities, and all the LIBs are sent to the recycling plant, where a set of recycling processes is available to recover materials from LIBs. Once the valuable materials from batteries are recovered, they are sold to customers. Note that, to simplify the model, battery waste or residues are considered products; however, all of them are sent to a single specific consumption point named “waste”. The following assumptions have been established to streamline the model and simplify its resolution: (1) A period of 1 year is considered; (2) Both collection centers and consumption points are predetermined; (3) Recycling processes, if the model selects it, are installed at the same geographical location, allowing the potential for multiple recycling processes in a single recycling plant; (4) The products obtained from the recycling processes are essentially the chemical elements that constitute the cathode of LIBs; (5) The quantity of LIBs available for recycling was established randomly; (6) The optimization model only considers pyrometallurgical and hydrometallurgical processes, direct recycling processes are not incorporated in this study.

To elucidate the model, various sets were defined as: (1) collection facilities, ; (2) types of batteries, ; (3) recycling processes, ; (4) products, ; (5) consumption points, . Subsequently, the model parameters are detailed in Table 1.

Table 1: Model parameters.

|  |  |  |
| --- | --- | --- |
| Symbol | Unit | Description |
|  | t | Recycling capacity of each recycling process . |
|  | US$ | Fixed operating cost of every recycling process in the recycling plant. |
|  | km | Distance between each collection center and the recycling plant. |
|  | km | Distance between the recycling plant and every consumption point . |
|  | tbattery-1 | Mass of the battery . |
|  | battery | The available quantity of battery at the collection center . |
|  | US$battery-1 | Sale price for the product in each consumption point  |
|  | - | The recovery efficiency of product in each recycling process . |
|  | - | Mass fraction of product that can be found in each battery . |
|  | US$t-1 | The unit cost of processing battery through the recycling process. |
|  | US$battery-1km-1 | Cost to transport each battery type from collection point to the recycling plant. |
|  | US$t-1km-1 | Cost to transport product from the recycling plant to each consumption point. |
|  | - | Maximum number of recycling processes that can be selected. |

The optimization model minimizes the total cost of the recycling network. To achieve this purpose, an objective function (OF) has been defined considering four terms: (1) cost of recycling, (2) cost of transporting batteries from collection facilities to the recycling plant, (3) cost of transporting products from recycling plant to consumption points, and (4) income from sales. The OF is shown in (1).

|  |  |
| --- | --- |
|  | (1) |

Moreover, the model has the following constraints:

|  |  |  |  |
| --- | --- | --- | --- |
|  | (2) |  | (3) |
|  | (4) |  | (5) |
|  | (6) |
|  | (7) |  | (8) |
|  | (9) |  | (10) |
|  | (11) |  | (12) |

Constraints (2) and (3) are logical expressions where is a sufficiently large number that does not impact the space solution of the model. Constraint (4) implies that all LIBs are being recycled. Constraint (5) establishes that the feedstock LIBs to the recycling plant cannot exceed its capacity. Constraint (6) is a key component of the optimization model because this constraint describes the transformation of LIBs to the products. Constraint (7) defines that all recovered materials are shipped to the consumption points. Finally, constraints (8) to (12) describe the domain of the decision variables.

* 1. Case study

A case study was generated to validate and apply the proposed model. The case study consists of 5 collection facilities, four recycling processes, three customers, and one waste disposal, as shown in Figure 1. Each process has the same maximum recycling capacity (6,000 t). The processes considered for this conceptual recycling network are based on available recycling technologies to date, encompassing pyrometallurgical and hydrometallurgical methods. Pyrometallurgical processes use high temperatures to recover materials. Generally, this process has three steps: roasting, smelting, and refining. It is important to note that pyrometallurgical processes do not recover lithium. Instead, all of the lithium is directed to the slag for further treatment with another recycling technology. Hydrometallurgical processes consist of separation by components: leaching, solvent extraction, and chemical precipitation. Both types of processes are highly used on an industrial scale. (Li et al., 2023). The literature indicates various processes and recycling products, such as alloys, metals, salts, and hydroxides (Dobó et al., 2023). Details on the recycling efficiency of each process for the respective products are only briefly discussed in the literature. For this reason, in this case study, the efficiencies are defined theoretically based on the type of process and its complexity. The recovery efficiency of these two recycling processes for each product was set to 0.7, except for the recovery of lithium using pyrometallurgical recycling, in which the value was set to 0. Two new recycling processes were introduced: Ad-Hydro, a more sophisticated version of a conventional hydrometallurgical process, and Pyro+Hydro, which combines pyrometallurgical and hydrometallurgical recycling methods. Therefore, given that the two processes mentioned above are more complex than conventional recycling processes the recovery efficiency was set to 0.9 for both processes and each type of product.



Figure 1: Conceptual recycling network.

Three types of conventional LIBs are considered for this model: NCA, NMC, and LFP. The quantity of LIBs available for recycling is shown in Table 2. The number of LIBs has been generated randomly within a range from 700 to 1,500 batteries. This range of spent lithium-ion batteries is determined based on the current production levels of small-scale recycling industries. Additionally, it considers the emerging lithium battery recycling market (Latini et al., 2022). In this case study, 14,498 LIBs were considered in total.

Additionally, considering that a LIB has 0.365 t unit mass (Wang et al., 2020), there are 5,292 t of LIBs available to recycle in this case study. Moreover, only Li, Co, Ni, and Mn are assumed to be recovered from the cathode material. Gaines et al. (2018) provided the typical mass fraction composition of each battery type. Using this information, the quantity of the mentioned elements for each kind of LIB can be calculated, as detailed in Table 3. The materials not recovered from LIBs are considered waste. The unit transportation cost for transporting batteries from every collection facility to the recycling plant is 8 US$⋅battery-1⋅km-1; furthermore, the unit transportation cost between the recycling plant and every consumption point is 0.12 US$⋅t-1⋅km-1 (Li et al., 2018).

On the other hand, Dai et al. (2019) developed a model for estimating the cost of recycling networks, which includes capital and operational costs. The unit recycling costs are detailed in Table 4. Note that the costs for ad-hydro and pyro+hydro are assumed as follows: the costs for ad-hydro are 10% higher than the conventional hydrometallurgical process cost, and the costs for pyro+hydro are calculated as the average of the hydrometallurgical and pyrometallurgical costs. The sale price of Li was defined as 22,769 US$⋅t-1, 33,420 US$⋅t-1 for Co, 18,284 US$⋅t-1 for Ni, and 5 US$⋅t-1 for Mn (Show The Planet Inc., 2023). On the other hand, products considered waste have a disposal cost of 111.12 US$⋅t-1; note that the recycling plant must pay to dispose of these waste materials (Reinhart et al., 2023). Two scenarios of this case study were considered to solve the model: the first solves the described model, while the second modifies the OF by eliminating the income for the sales term.

|  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |  |
| --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- | --- |
| Table 2: Available quantity of each type of battery at each collection center (number of batteries).

|  |  |  |  |
| --- | --- | --- | --- |
|  | NMC | NCA | LFP |
|  | 1,269 | 701 | 901 |
|  | 946 | 1,032 | 767 |
|  | 1,122 | 870 | 1,481 |
|  | 866 | 882 | 801 |
|  | 1,363 | 783 | 714 |

 | Table 3: Mass fraction of LIBs components.

|  |  |  |  |
| --- | --- | --- | --- |
| Products | NMC | NCA | LPF |
| Li | 0.03 | 0.02 | 0.01 |
| Co | 0.04 | 0.03 | 0.00 |
| Ni | 0.11 | 0.16 | 0.00 |
| Mn | 0.04 | 0.00 | 0.00 |
| Waste | 0.78 | 0.79 | 0.99 |

 |

Table 4: Unit recycling cost, including capital and operation cost (US$battery-1).

|  |  |  |  |  |
| --- | --- | --- | --- | --- |
| LIB | Pyro | Hydro | Ad-Hydro | Pyro+Hydro |
| NMC | 2,229 | 2,118 | 2,329 | 2,174 |
| NCA | 2,246 | 2,122 | 2,335 | 2,184 |
| LFP | 1,166 | 1,235 | 1,359 | 1,201 |

* 1. Results

The results are summarized in Table 5. In both scenarios, the cost of recycling comprises 57% of the total cost. Furthermore, considering the data defined in the case study, there is no profit even with the revenue obtained from the sales of the products. The sales amortize only 28% of the total costs. In scenario B, the model chose the cheapest option for recycling, even if it meant recovering less material from the LIBs.

On the other hand, in scenario A, the model chose the best option, recovering the maximum amount of material while using less capital. The recovered materials are sent to the nearest consumption point. In the optimization model, as previously mentioned, only the cathode of LIBs is recycled; therefore, a considerable quantity of materials are not processed; only 12% of the materials that comprise LIBs are recovered. However, it is valuable to highlight that the model may represent several scenarios besides those shown in this document. Further research may be performed by considering scenarios representing potential realizations of public policies, innovation projects, technology development, private initiatives, and cultural changes.

Nevertheless, the proposed model represents a valuable tool for designing the Li-ion batteries recycling supply chain network. Additionally, it is relevant to consider potential scenarios regarding the final destination of the waste generated through all the processes. This is crucial considering the low percentage recuperated from the processed batteries and is highly dependent on the potential scenarios recently mentioned.

Table 5: Case study results

|  |  |  |  |
| --- | --- | --- | --- |
| Scenario | RN costs | Recycling process selected | Waste |
| A | 47.0 MM USD | Pyro+Hydro | 4,577 t |
| B | 46.5 MM USD | Hydro | 4,736 t |

* 1. Conclusions

An optimization model was developed to design recycling networks of LIBs. Based on the data provided, the results only apply to the proposed conceptual recycling network, considering the number of battery types and recovery efficiency. However, the optimization model can be used to assess what recycling process is optimal according to the needs of the user. Continuing research on the recovery efficiencies of today’s industrial-scale recycling processes is highly recommended, as there is a lack of information from a technical and scientific perspective. From a chemical and industrial engineering perspective, it is relevant to focus on designing recycling networks that can manage and process all components of LIBs. The proposed model represents a valuable tool for managers and policymakers considering the potentiality of evaluating several potential scenarios regarding potential developments and policies. Consequently, the results obtained are highly dependent on the possible scenarios. Still, it enables considering the consequences of the decisions and developments impacting technologies, efficiencies, and other relevant aspects for designing the Li-ion batteries recycling supply chain network.

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