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Research article

Designing profitable supply chains for lithium-ion battery recycling in the United States

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Abstract: Recycling spent lithium-ion batteries (LIBs) has attracted lots of attention recently, due to the increasing demand for critical materials contained in LIBs, putting high pressure on their geological reserves. We evaluated the potential of bioleaching technology as a sustainable solution for recycling spent LIBs to help inform decision-making processes for stakeholders involved in LIB recycling supply chains. A supply chain model was developed to include required upstream processes with the objective of maximizing economic feasibility of LIB recycling through the technology. The model has been applied to the U.S. and an optimal supply chain configuration was identified, considering the major factors affecting the economic viability of the technology. The net present value of the supply chain was estimated to be $18.4 billion for operating over 10 years, achieving the maximum processing capacity of 900,000 tons of black mass per year. The economic viability of the technology was identified to be highly sensitive to the cost associated with purchasing black mass, which accounted for more than 60% of the total supply chain cost. The breakeven price of black mass was identified as $8.7/kg over which the supply chain was not economically sustainable. Additionally, we examined the non-cooperative scenarios where each tier tries to maximize its own profit to demonstrate how the overall profitability of the supply chain changes with different pricing strategies of sortation facilities and acid producers. We estimated that the maximum prices of non-recyclable paper and acid that the supply chain could tolerate were $0.89/kg and $8.5/kg, respectively, beyond which the supply chain was no longer sustainable.

Keywords: lithium-ion battery; circular economy; bioleaching; critical materials recovery; non-cooperative supply chain

1. Introduction

Lithium-ion batteries (LIBs) have become a crucial part of clean energy. The rapid growth of electric vehicles (EVs) and consumer electronics has led to a surge in the production and consumption of LIBs. However, the disposal of spent LIBs has raised serious environmental concerns due to its hazardous contents, including heavy metals and toxic chemicals. Therefore, with the growing demand for LIBs, it is crucial to develop safe and sustainable disposal methods that address environmental concerns. The United States, in particular, faces a growing challenge of managing the end-of-life (EOL) LIBs, with projections indicating that over 2.6 million tons of batteries per year will be available recycling in North America by 2030 [1]. Moreover, recycling LIBs can help in covering the demand for valuable critical materials, including cobalt, nickel, and lithium, which are predominantly mined outside of the United States. The recycling of LIBs can help reduce the US dependency on other countries, increasing its energy security and creating opportunities for domestic production of critical materials.

LIBs are typically recycled using pyrometallurgical and hydrometallurgical technologies [2]. The pyrometallurgical process involves heating the batteries to high temperatures to melt and separate the metal components in an alloy form. The hydrometallurgical process requires further pre-processing of spent batteries to extract cathode active materials (CAM) [3]. The metal components in CAM (or alloy from pyro-process) will be dissolved in acids, followed by precipitation and recovery. These technologies are effective in separating metals from LIBs; however, they have several drawbacks, including high energy consumption, high emissions of greenhouse gases, and the production of hazardous waste [4,5]. Bioleaching technology has emerged as a promising alternative for LIB recycling, offering environmental benefits over traditional energy-intensive recycling technologies and a high level of economic feasibility [6]. Bioleaching is a microbial process that utilizes microorganisms that produce organic and/or inorganic acids to dissolve metal components from LIBs [7]. The bacteria used in bioleaching can be sourced from natural environments, making the process cost-effective and sustainable. A recent study by Idaho National Laboratory and the University of Arizona explored the economic feasibility of using *Gluconobacter oxydans* to produce organic acids from municipal solid waste (MSW) such as non-recyclable paper [8]. The ability to recycle waste LIBs through bioleaching technology, which utilizes some waste streams in MSW for lixiviant production, makes the technology more economically and environmentally attractive.

Recycling of waste LIBs in the United States is a promising alternative to sending LIB black mass (i.e., LIB cathode containing powder) overseas for recovering valuable metals, from both economic and environmental perspectives. However, the low economic return has been a significant challenge to the feasibility of domestic LIB recycling [9]. High economic viability of bioleaching technology in value recovery from LIB black mass could promote sustainable recycling of spent LIBs in the United States, if an optimal supply chain configuration could be found to connect all the required upstream tiers. In addition, a well-designed recycling network can improve the efficiency of waste collection and transportation, reduce costs, and ensure that LIB waste is directed to the appropriate recycling facilities.

Designing a network for LIB recycling using bioleaching technology in the United States is not without its challenges. The fact that the technology is relatively new and there is a lack of necessary infrastructure is one of the greatest barriers. Thus, the economic viability of bioleaching technology for LIB recycling is uncertain and requires detailed investment plan for necessary equipment and infrastructure. Additionally, the market demand for recycled LIB materials is developing, which may affect the profitability of the process. To overcome these challenges, collaboration among stakeholders is crucial. The LIB black mass producers, the MSW sortation facilities, organic acid producers, and bioleaching facilities must work together to ensure an efficient flow of materials through a comprehensive recycling network.

In this article, we aim to provide insights into the design of an optimal network for LIB recycling using bioleaching technology in the US market to maximize economic sustainability. The network includes the required upstream processes to separate non-recyclable paper from MSW to produce organic acid for waste LIB bioleaching. Particularly, we will optimize the main variables, such as the location of facilities, their operating capacities, and transportation flows to maximize the net present value (NPV) of the entire supply chain and discuss the key factors affecting the network sustainability. By doing so, this study contributes to the development of a sustainable LIB recycling supply chain that can help meet the increasing demand for critical materials in the United States.

2. Literature review

The management of EOL LIBs from EVs has become a critical concern due to the increasing demand for sustainable transportation. Moreover, LIBs are class 9 hazardous materials and pose a risk to human health and the environment. Therefore, it is crucial to safely collect and transport the EOL products to downstream processors for recycling [2]. This literature review aims to summarize recent research on developing quantitative models for LIB recycling network.

Wang et al. (2020) [10] proposed an optimal design of an EV battery recycling network from the perspective of EV manufacturers. Their model focused on reusing, remanufacturing, and recycling spent LIBs. The authors identified transportation costs, the number of used EV batteries, and carbon tax as the prime factors affecting the optimal design of the recycling network. The results showed that using the hierarchical design of the recycling network, including regional and central facilities, could reduce the total cost and environmental impact. Hoyer et al. (2015) [11] presented a technology and capacity planning model for the recycling of Li-ion EV batteries in Germany. Their model analyzed the optimal capacity of recycling plants, investment decisions, and technological advances. The authors proposed a sequential deployment of recycling plants over time to deal with the uncertain and dynamic LIB market. The strategy resulted in a higher NPV when compared to the plan of establishing all the plants at the beginning of the planning horizon. Tadaros et al. (2020) [12] applied the same sequential deployment strategy in their reverse logistics model to accommodate the increasing volume of spent LIBs. Their model focused on location and network design for reverse logistics of LIBs in Sweden. The authors identified the optimal number and location of collection and recycling facilities and transportation routes to minimize the total cost of the network. Hendrickson et al. (2015) [13] combined life-cycle assessment (LCA) and geographic information systems (GIS) to analyze the energy consumption, greenhouse gas (GHG) emissions, water use, and criteria air pollutant emissions from EOL infrastructure networks for recycling LIBs in California. Using economic and environmental criteria, GIS modeling revealed optimal locations for battery dismantling and recycling facilities for in-state and out-of-state recycling scenarios. The results showed that material recovery from pyrometallurgy can offset environmental burdens associated with LIB production, namely a 6%–56% reduction in primary energy demand and 23% reduction in GHG emissions, when compared to virgin production. Gonzales-Calienes et al. (2022) [14] also utilized a combination of GIS, material flow analysis, and LCA to pinpoint optimal sites for dismantling hubs and complete recycling facilities, considering both economic and environmental factors. Rosenberg et al. (2023) [15] developed a dynamic network design model with capacity expansions for EOL traction battery recycling. The authors applied the model to a case study of an original equipment manufacturer (OEM) in Germany and analyzed the optimal location of the recycling plant, the network structure, and the investment decisions. Their results showed that investing in multiple recycling facilities with different capacities can lead to cost savings and reduce the risk of supply disruptions. Finally, Yükseltürk et al. (2020) [16] proposed a model for collection center location for EOL EV batteries using fleet size forecast. Their model analyzed the optimal location of the collection centers and the required fleet size to collect EOL batteries from different regions in Germany. The authors considered different scenarios with different collection rates and showed that their model could help decision-makers optimize the collection network and reduce the transportation cost. A summary of these studies is provided in Table 1. These studies proposed quantitative models for designing the reverse logistics network for LIB recycling. They analyzed the optimal locations of collection and recycling facilities, transportation routes, and the investment decisions required to minimize the total cost and environmental impact of the network. The results provide insights for policymakers and decision-makers to design efficient and sustainable recycling networks for EOL LIBs.

Besides strategic decision making, researchers have delved into evaluating policy efficacy and supply chain dynamics through game theory, particularly focusing on the Stackelberg game model. Zhang et al. (2023) [17] investigated the impact of government incentives, such as subsidies and deposits, on Chinese stakeholders using Stackelberg models. They discovered that these incentives effectively enhanced recycling rates, economic gains, and environmental outcomes within closed-loop supply chains. Similarly, Zhao et al. (2022) [18] applied the Stackelberg game to analyze pricing strategies for EV batteries, proposing subsidies for vehicle manufacturers and battery producers in China to promote remanufacturing and recyclability. Recent studies have also employed the Stackelberg modeling approach to examine the competitive dynamics of LIB reverse logistics networks. Lin et al. (2023) [19] assessed various government regulatory schemes through Stackelberg games between EV manufacturers and retailers and found that policies providing rewards or penalties based on recycling performance yielded the highest recycling rates. Gu et al. (2023) [20] demonstrated that government penalties effectively encouraged battery recycling participation and cooperation among supply chain entities in manufacturer-retailer Stackelberg models. Furthermore, Li et al. (2023) [21] utilized evolutionary game theory involving vehicle producers, battery manufacturers, and recycling firms, emphasizing the importance of subsidies and digitalization in establishing sustainable closed-loop supply chains. They argued that without these elements, profitable battery reclamation would not naturally occur.

Table 1. Reverse logistics optimization for spent LIBs (Adapted from Alipanah et al. (2021) [2] with permission. Copyright (2022) AIMS Press). "LCA" stands for life cycle analysis.

Reference	Decision variables	Technology	Objective function	Modeling approach	Solution method	Sustainability dimension	Location
[10]	Optimizing facility locations and material flows for three alternative strategies: recycling, disposal, and remanufacturing EOL LIBs	-	Minimizing operations cost and CO_2 emissions	Mixed integer linear programming	Genetic algorithm	Economic and Environmental	China
[11]	Determining the optimal investment plan for a sequential deployment of LIB recycling plants	-	Maximizing net present value of cash flow	Mixed integer linear programming	CPLEX software	Economic	Germany
[12]	Optimizing facility location, transportation quantity, and allocation of demand zones to each facility	-	Minimizing total annual cost	Mixed integer linear programming	Unspecified	Economic	Sweden
[13]	Optimizing facility location	-	Minimizing capital and transportation costs	LCA & Geographic information system	Unspecified	Economic and Environmental	California
[16]	Optimal location of recollection centers, Assignment of operation zones to a selected recollection center	-	Minimize the sum of the transportation time of all batteries	Mixed integer linear programming	Gurobi	-	Germany
[15]	Optimizing facility location with its capacity level, transportation quantity	-	Minimize total cost	Mixed integer linear programming	CPLEX	Economic	Germany
This study	Optimizing facility location, processing capacity, and transportation flow	Bioleaching	Maximizing net present value	Mixed integer linear programming	CPLEX	Economic	United States

2.1. Literature gap and contribution

Despite the abovementioned publications in reverse logistics network design for LIB recycling, there are several research gaps that need to be addressed. Our research contributes to the literature in the following aspects.

(1) Our research is tailored to a LIB supply chain based on advanced sortation and bioleaching technologies, which are promising alternatives to the current business practices [8,22]. None of the literature, to the best of our knowledge, has discussed the technological impacts on supply chain decisions.

(2) Poor economic feasibility has been a major challenge of recycling LIBs in the United States, and most LIB wastes are exported to other countries. This challenge highlights the necessity of configuring an optimal supply chain for economical recycling of LIBs in the United States.

(3) Our research takes into account the changing material composition and availability of waste LIBs and recovered material prices in the future, which can significantly impact the design of the recycling network. None of the previous studies in the abovementioned literature have explicitly addressed this issue.

(4) We compare the performance of a vertically integrated supply chain with a non-cooperative supply chain in our model to illustrate the benefits of an integrated supply chain, which has not been explored in previous research.

3. Problem statement

Material recycling facilities (MRFs) are specialized in managing different types of waste materials from residential and commercial sources. These facilities play an important role in diverting waste from landfills. The recycling process at an MRF includes initial inspection, separation, and processing of materials into marketable commodities such as bales of plastic, paper, and metal. Recent technological advancements have enhanced the efficiency and effectiveness of MRFs, enabling better material recovery rates and enhanced sortation accuracy. AMP Robotics [23], for instance, is developing automated solutions for MRFs based on artificial intelligence (AI) and machine learning of sensor and camera data in order to improve materials characterization and sortation efficiency and reduce contamination.

Non-recyclable paper obtained from MRFs could be employed as a nutrient source to produce a biolixiviant through fermentation of *Gluconobacter oxydans*. The biolixiviant includes gluconic acid and xylonic acid and is utilized to leach waste LIB materials commonly referred to as black mass [8]. The process of leaching is carried out in the presence of Fe^{2+}, a reducing agent. The outputs of the bioleaching process are solubilized cobalt, lithium, nickel, and manganese that will be subject to downstream separation and precipitation processes.

We propose a novel supply chain in which advanced sortation technologies are incorporated to some of the existing MRFs to separate non-recyclable paper from other waste streams. Separated non-recyclable paper is subsequently transported to the facilities of organic acid producers, where it is used to produce organic acids. The organic acid is delivered to bioleaching facilities, where LIB black mass is bioleached to recover critical metals. The supply chain facilities and material flows are depicted in Figure 1.

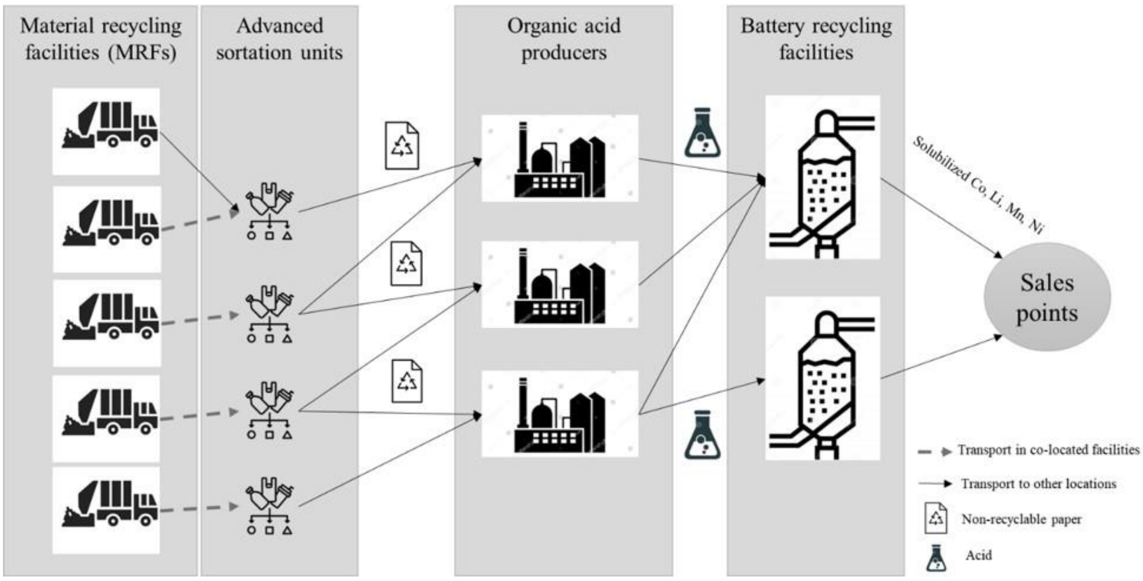

Figure 1. Material flow diagram in the supply chain for waste LIB bioleaching.

As we consider sorting MSW as the starting point of the supply chain, the decision must be made regarding the selection of MRFs that should be equipped with advanced sortation technologies. To optimize supply chain economics overall, acid producers and bioleaching facilities try to identify their optimal location and operational capacity. Given the projected increase in the amount of spent lithium-ion batteries (LIBs) in the coming decade, it is expected that facilities will need to consistently increase their processing capacities throughout the planning period.

3.1. Bioleaching supply chain model assumptions

Bioleaching process. This paper incorporates the main assumptions for the bioleaching process from a recent study which showed the successful recovery of critical metals through gluconic acid based bioleaching of LIB black mass based on techno-economic analysis modeling of an industrial scale operation [22]. The study conducted by Alipanah et al., 2023, sourced the black mass from Retriev Technologies (now Cirba Solutions in Trail, British Columbia, Canada), revealing the black mass weight composition to be 2.3% lithium, 19.3% cobalt, 4.5% manganese, and 5.6% nickel. According to Ekberg C (2015) [24], spent LIBs undergo a series of mechanical treatments such as multistage crushing, sieving, magnetic separation, and fine crushing to remove steel casing and current collectors to produce black mass. The black mass was leached at a 2.5% solid to liquid ratio, at 55 ℃ temperature over a duration of 30 hours with a biolixiviant containing 75 mM gluconic acid. Iron(II) sulfate was utilized as a reducing agent to facilitate the dissolution of metals. The average leaching efficiencies of the critical metals were reported as 100% for both lithium and manganese, 84% for cobalt, and 81% for nickel.

Economic estimation. The techno-economic assessment by Alipanah et al. [22] for an industrial implementation of LIB black mass bioleaching is utilized as the foundation for the cost and revenue estimations of the bioleaching facilities for the current study. The model assumed a bioleaching plant in the US capable of recycling 10,000 tons of LIB black mass per year. All the relevant bioleaching costs are translated into fixed cost (e.g., costs of leaching tank, land, equipment installation) and operating cost (e.g., costs of material, utility, waste management, labor). Considering that the products of bioleaching are in the form of solubilized metals rather than ≥99% pure metal salts sold in the market, a discount factor of 30% is applied for calculating the revenue (i.e., bioleached metals are assumed to be sold at 70% of their market price). Detailed costs assumptions are provided in section 4 for a case study.

3.2. Mathematical formulation

In this section, a mixed-integer linear programming (MILP) model is developed for identifying the optimal facility locations, processing capacities, and transportation flows to maximize NPV of the overall supply chain described in Figure 1 over ten years of planning horizon. Constant parameters are denoted by capital letters, and decision variables are denoted in lower cases.

Notations

$m \in M$: Index for MRFs.
$s \in S$: Index for sortation facilities.
$a \in A$: Index for acid producers.
$b \in B$: Index for bioleaching facilities.
$t \in T$: Index for time horizon.

Parameters

FC_s^{Sort}: Fixed cost for equipping MRF facility *s* with advanced sortation technology.
FC^{Acid}: Fixed cost for building acid producer facility.
FC^{Bio}: Fixed cost for building bioleaching facility.
UF^{Acid}: Unit increase in the fixed cost of acid producer with a unit increase in processing capacity.
UF^{Bio}: Unit increase in the fixed cost of bioleaching facility with a unit increase in processing capacity.
OC_s^{Sort}: Unit operating cost of advanced sortation facility *s*.
OC_a^{Acid}: Unit operating cost of organic acid producer *a*.
OC_b^{Bio}: Unit operating cost of bioleaching facility *b*.
Tr_{ms}^{Waste}: Unit transportation cost of transporting waste from MRF *m* to sortation facility *s*.
Tr_{sa}^{OWaste}: Unit transportation cost of transporting non-recyclable paper from sortation facility *s* to acid producer *a*.
Tr_{ab}^{Acid}: Unit transportation cost of transporting acid from acid producer *a* to bioleaching facility *b*.
Q_s^{MRF}: Capacity of sortation facility *s*.
Q^{Acid_b}: Base processing capacity of acid producers.
Q^{Acid_m}: Maximum processing capacity of acid producers.
Q^{Bio_b}: Base processing capacity of bioleaching facilities.
Q^{Bio_m}: Maximum processing capacity of bioleaching facilities.
Con^{Pwaste}: Portion of non-recyclable paper in MSW.
Eff^{Sort}: Sortation facilities' efficiency.
Con^{Acid}: Conversion rate of non-recyclable paper to organic acid.
Eff^{Acid}: Efficiency of acid producers.
UA: Unit acid consumption for bioleaching of unit black mass.
R^{Co}: Recovery rate of cobalt in the bioleaching facilities.
R^{Li}: Recovery rate of lithium in the bioleaching facilities.
R^{Ni}: Recovery rate of nickel in the bioleaching facilities.

DR: Discount rate on the price of bioleaching output metals.
$P^{non-paper}$: Unit price of non-paper separated in sortation facilities.
P^{bm}: Unit price of black mass.
P^{Co}: Unit price of cobalt.
P^{Li}: Unit price of lithium.
P^{Ni}: Unit price of nickel.
C^{co}: Cobalt content in black mass.
C^{Li}: Lithium content in black mass.
C^{Ni}: Nickel content in black mass.

Decision variables

$y_{st} = \begin{cases} 1 \\ 0 \end{cases}$ if sortation facility s is built at time t.

$x_{st} = \begin{cases} 1 \\ 0 \end{cases}$ if sortation facility s is operational during time t.

$y_{at} = \begin{cases} 1 \\ 0 \end{cases}$ if acid producer a is built at time t.

$x_{at} = \begin{cases} 1 \\ 0 \end{cases}$ if acid producer a is operational during time t.

$y_{bt} = \begin{cases} 1 \\ 0 \end{cases}$ if bioleaching facility b is built at time t.

$x_{bt} = \begin{cases} 1 \\ 0 \end{cases}$ if bioleaching facility b is operational during time t.

q_{st}: Amount of processed waste in sortation facility s during time t.
cap_{at}: Non-recyclable paper processing capacity of acid producer a during time t.
q_{mst}: Amount of waste transported from MRF m to sortation facility s during time t.
q_{sat}: Amount of non-recyclable paper transported from sortation s to acid producer a during time t.
q_{abt}: Amount of acid transported from acid producer a to bioleaching facility b during time t.
cap_{bt}: Black mass processing capacity of bioleaching facility b during time t.
ex_{at}: Extra added capacity to acid producer a at time t.
ex_{bt}: Extra added capacity to bioleaching factory b at time t.

3.2.1. Optimization model

The objective function of the supply chain is to maximize the total profit during the assumed planning horizon. To consider the time value of investments, the supply chain's profit is translated to NPV which is shown in Eq 1. MARR denotes the minimum acceptable rate of return.

Objective function:

$$Max\ NPV = \sum_t \frac{Revenue_t - Cost_t}{(1 + MARR)^t} \tag{1}$$

Two sources of revenue are assumed: (1) selling the separated wastes, excluding paper, in MRFs by the advanced sortation facilities (Eq 2) and (2) selling solubilized metals from bioleaching facilities (Eq 3).

$$\sum_t \sum_s q_{st} . (1 - Con^{Pwaste}) . P^{non-paper} \tag{2}$$

$$\sum_t \sum_b cap_{bt} . (C^{Co} . R^{Co} . P^{Co} + C^{Li} . R^{Li} . P^{Li} + C^{Ni} . R^{Ni} . P^{Ni}) . DR \tag{3}$$

Three types of costs are assumed in the supply chain, setup cost of each facility when it is constructed (Eq 4), operating cost (Eq 5), and transportation costs (Eq 6). Furthermore, acid producers and bioleaching facilities are assumed to be able to expand their processing capacity each year.

$$\sum_t (\sum_s (y_{st} . FC_s^{Sort}) + \sum_a (y_{at} . FC^{Acid} + \sum_{i=1}^{t} UF^{Acid} . ex_{ai}) + \sum_b (y_{bt} . FC^{Bio} + \sum_{i=1}^{t} UF^{Bio} . ex_{bi})) \tag{4}$$

$$\sum_t \sum_s OC_s^{Sort} . q_{st} + \sum_t \sum_a OC_a^{Acid} . cap_{at} + \sum_t \sum_b (OC_b^{Bio} + P^{bm}) . cap_{bt} \tag{5}$$

$$\sum_t (\sum_m \sum_s Tr_{ms}^{Waste} . q_{mst} + \sum_s \sum_a Tr_{sa}^{OWaste} . q_{sat} + \sum_a \sum_b Tr_{ab}^{Acid} . q_{abt}) \tag{6}$$

The objective function is subjected to the following constraints. Eqs 7–12 ensure that a facility is operational at time *t* if it was operational at time *t-1*, or it was built at time *t*. These constraints also prevent a facility from being built multiple times.

$$x_{st} = x_{st-1} + y_{st} , \quad \forall s, t/1 \tag{7}$$

$$x_{s,1} = y_{s,1} \tag{8}$$

$$x_{at} = x_{at-1} + y_{at} , \quad \forall a, t/1 \tag{9}$$

$$x_{a,1} = y_{a,1} \tag{10}$$

$$x_{bt} = x_{bt-1} + y_{bt} , \quad \forall b, t/1 \tag{11}$$

$$x_{b,1} = y_{b,1} \tag{12}$$

Equations 13–17 constrain the operational capacity of facilities to their maximum capacities.

$$q_{st} \leq Q_s^{Sort} . x_{st} , \quad \forall s, t \tag{13}$$

$$cap_{at} \leq Q_b^{Acid-b} . x_{at} + \sum_{i=1}^{t} ex_{ai} , \quad \forall a, t \tag{14}$$

$$\sum_{i=1}^{t} ex_{ai} \le Q^{Acid\_m}.x_{at}, \quad \forall a, t \tag{15}$$

$$cap_{bt} \le Q^{Bio\_b}.x_{bt} + \sum_{i=1}^{t} ex_{bi}, \ \forall b, t \tag{16}$$

$$\sum_{i=1}^{t} ex_{bi} \le Q^{Bio\_m}.x_{bt}, \quad \forall b, t \tag{17}$$

The amount of received materials should be equal to the amount of processed material (Eqs 18–20).

$$\sum_{m} q_{mst} = q_{st} \quad \forall s, t \tag{18}$$

$$\sum_{s} q_{sat} = cap_{at}, \forall a, t \tag{19}$$

$$\sum_{a} q_{abt} = UA.cap_{bt}, \forall b, t \tag{20}$$

Equation 21 ensures that amount of transferred MSW from a MRF should be within the actual capacity of that MRF. Eqs 22 and 23 show that amount of separated non-recyclable paper and produced acid are proportional to amount of processed MSW and organic waste, respectively. Eq 22 enables sorting facilities to sort more waste than acid producers require in order to generate a higher revenue from the direct sale of sorted waste.

$$\sum_{s} q_{mst} \le Q_m^{MRF}, \quad \forall m, t \tag{21}$$

$$\sum_{a} q_{sat} \le q_{st}.Con^{Pwaste}.Eff^{Sort}, \quad \forall s, t \tag{22}$$

$$\sum_{b} q_{abt} = cap_{at}.Con^{acid}.Eff^{Acid}, \quad \forall a, t \tag{23}$$

Equation 24 ensures that amount of processed black mass in all bioleaching facilities should be less than black mass availability at the time *t*. Eq 25 ensures non-negativity of the variables.

$$\sum_{b} cap_{bt} \le D_t^{black\,mass}, \forall t \tag{24}$$

$$q_{st}, cap_{at}, q_{sat}, q_{abt}, cap_{bt} \ge 0, y, x \in \{0,1\} \tag{25}$$

As the problem is a traditional MILP, it could be solved using the available commercial solvers. Therefore, IBM ILOG CPLEX package was used in Python to solve the modeled problem in a computer with a core-i7 processor and 16 GB of RAM.

4. Case study

The proposed MILP model was applied to design an optimal supply chain for LIB recycling in the US market.

(1) The volume of black mass to recycle in the US was estimated based on the data from Li-Cycle [1] for the next decade, which increased from 300,000 tonnes in year 1 to 1,500,000 tonnes in year 10. The materials include EOL LIBs from consumer electronics and EVs as well as LIB manufacturing scraps.

(2) The location and capacity of MRFs in the US were identified from ZeeMap [25], including 68 major MRFs across different states. These MRFs were considered as candidates for co-locating advanced sortation facilities.

(3) Various companies announced their plan of developing LIB recycling facilities in California, Texas, New York, Ohio, Arizona, Tennessee, Indiana, Michigan, and Nevada [26], so these states were considered as candidate locations for constructing bioleaching facilities. Downstream processes (i.e., sales points) of bioleaching were assumed to have the same candidate locations to minimize transportation of a large volume of bioleached materials.

(4) The study assumed that the sales price of recovered metals would increase by approximately 5% each year over the next decade [26].

(5) The solubilized metals from bioleaching were assumed to be sold at a 30% discounted price of their corresponding metal prices to account for the downstream processing costs [22].

(6) The operating cost of each facility varies according to the electricity cost rate of each state [27].

(7) The distances between facilities were estimated based on the actual road distances calculated by Google Maps [28].

(8) The study also incorporated changes in critical material composition in LIB cathodes over the next decade, reflecting the trend of decreasing cobalt and increasing nickel content [29].

Table 2 provides a summary of the key input parameters used in the model.

Table 2. Key parameters and values used in LIB recycling supply chain model.

Parameter	Value	Unit	Data source
Portion of wastepaper in MSW	24%		[30]
Mass conversion rate of non-recyclable paper into acid	55%		[8]
Sortation efficiency	95%		Our assumption
Acid producer efficiency	95%		[8]
Black mass price	$3.5	Per kg	[22,31]
Transportation cost	$0.0001	Per kg-km	[31]
Fixed cost of establishing an advanced sortation facility with capacity of 63,000 t/y of MSW. (Fixed cost of sortation facilities varies proportional to their capacity)	$800,000	Per facility	Our estimation
Fixed cost of constructing an acid producing facility (capacity: 20,000 t/y of wastepaper)	$60,000,000	Per facility	Estimated from [8]
Fixed cost of constructing a bioleaching facility (capacity: 10,000 t/y of black mass)	$44,000,000	Per facility	Estimated from [22]
Capacity expansion cost for acid producers	$0.29	Per kg paper	Estimated from [8]
Capacity expansion cost for bioleaching facilities	$0.32	Per kg	Estimated from [22]
Maximum processing capacity of an acid production facility	200,000	t (paper)/y	Our assumption

Continued on next page

Parameter	Value	Unit	Data source
Maximum processing capacity of a bioleaching facility	100,000	t (black mass)/y	Our assumption
Price of cobalt	$55.7	Per kg	2018–2022 average [32]
Price of lithium carbonate	$17.1	Per kg	2018–2022 average [32]
Price of nickel	$16.9	Per kg	2018–2022 average [32]
Black mass weight to battery weight	51%	Per pack	[22]
MARR	8%	Per year	[22]
Project life	10	Years	Our assumption

4.1. Case study results

The optimal supply chain configuration is shown in Figure 2a for year 1 and in Figure 2b for year 10 of the project. The network is more concentrated on the west coast, east coast, and Texas because these are the areas with the highest population density and the largest MRFs to feed the supply chain with enough non-recyclable paper. The required non-recyclable paper stream for acid producers was fulfilled from the closest neighboring MRFs to minimize the transportation cost. It is important to note that the decision regarding the establishment of acid production facilities was not solely influenced by the operational expenses associated with a given location. Rather, factors such as the presence of MRFs capable of generating an adequate supply of non-recyclable paper, as well as the transportation costs associated with both the non-recyclable paper and the produced acid to bioleaching facilities, were also taken into account. In fact, the model considered the trade-off between transportation cost and operational cost to build new facilities (e.g., building acid producer facility in Oregon vs. co-locating acid producer and bioleaching facilities in Texas).

To maximize the NPV, the optimal solution suggested opening bioleaching facilities in all nine candidate locations in California, Texas, New York, Ohio, Michigan, Arizona, Tennessee, Indiana, and Nevada. The required organic acids were supplied through six acid production facilities in Texas, New York, Ohio, Arizona, Indiana, and Oregon. All 68 MRFs around the US were selected for adopting advanced sortation technologies to provide the required non-recyclable paper stream to acid producer facilities. In fact, there are more non-recyclable paper available to produce organic acid needed for LIB recycling, and as LIB recycling increases, there is room to grow and utilize more MRFs and sortation facilities for bioleaching.

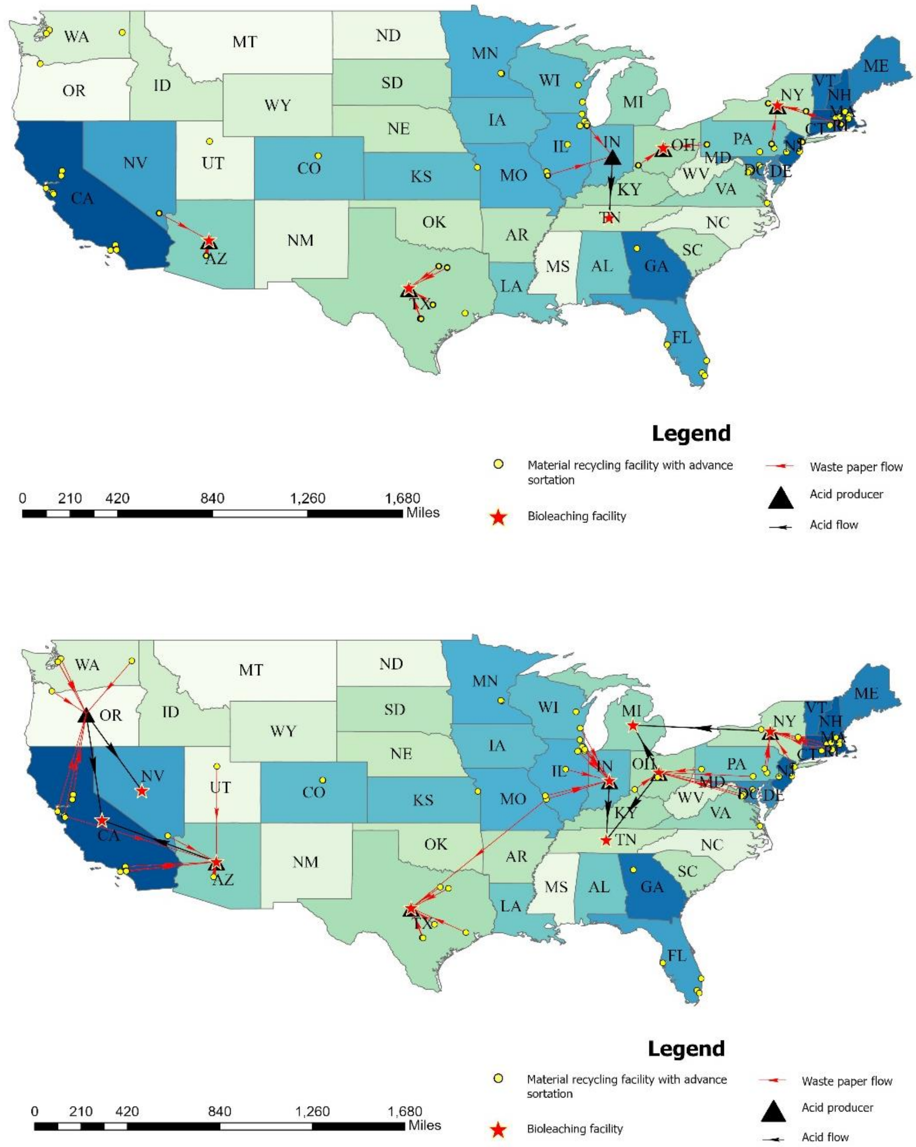

Figure 2. Geographic representation of optimal supply chain configuration for recycling LIB black mass through bioleaching and the transportation flows in the first year (a) and last year (b) of the planning horizon. Darker color shows higher electricity cost.

Figure 3 illustrates the gradual increase in the number of acid producers and their operating capacities and Figure 4 illustrates those of bioleaching facilities. The supply chain starts with 5 acid producing facilities in Texas, New York, Ohio, Arizona, and Indiana to meet the demand of 5 bioleaching facilities in Texas, New York, Ohio, Arizona, and Tennessee. As the amount of LIB black mass increased in the following year, the model suggested opening another bioleaching facility in Nevada and another acid producer facility in Oregon. With a jump in the availability of black mass in year 3, the model suggested to open two new bioleaching facilities in Indiana and Michigan. Then each facility would increase their processing capacities, as a response to increasing black mass availability and recycling volume.

This optimized supply chain could recycle over 13 million tons of waste LIBs and 8 million tons of non-recyclable paper over the next 10 years, leading to an estimated NPV of $18.4 billion.

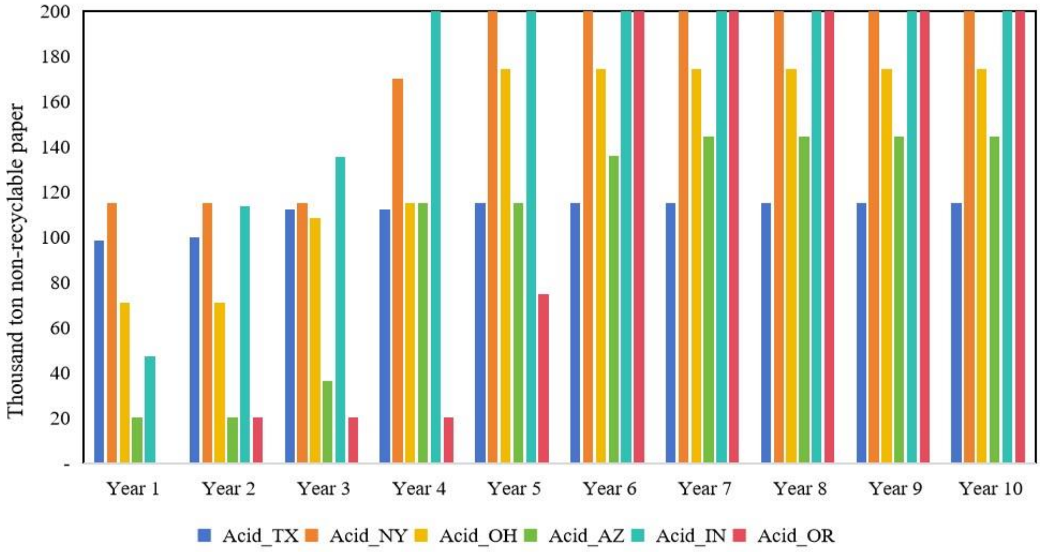

Figure 3. Gradual increase in processing capacity of the selected acid producer facilities over the 10 years planning horizon.

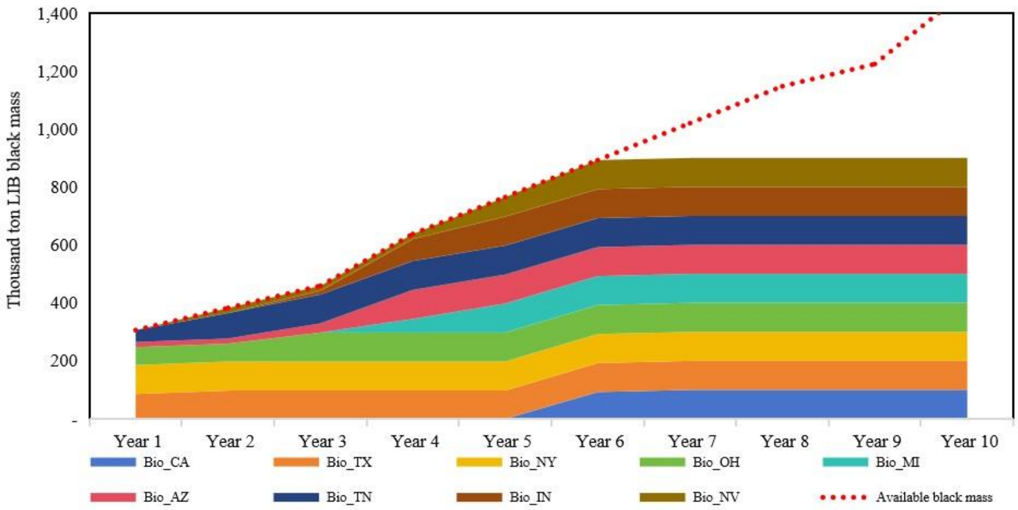

Figure 4. Contribution of each bioleaching facility in processing available black mass over the planning horizon. Red dashed line represents the projected availability of the LIB black mass in the United States from manufacturing scraps, consumer electronics, and electric vehicles.

5. Scenario analysis

As indicated in a previous study [22], the economic viability of adopting bioleaching technology for value recovery from LIB is not only related to the abovementioned decision variables but it is also sensitive to external factors. In this section, the focus is on two factors: the purchasing price of black mass and the sustainability of the supply chain under non-cooperative situations.

5.1. Black mass purchase price

The cost of purchasing black mass constitutes a significant portion of the total supply chain cost, with more than 60% attributed to this factor. Therefore, it is crucial to assess the supply chain's sustainability in the face of changing black mass prices. The base case assumes a purchasing price of around $3.5/kg, as reported by [26]. However, Wang and Yu (2021) [33] estimated that the cost of black mass preparation was around $5/kg. To evaluate the effect of increasing black mass prices, a range of scenarios from the base case to the edge case (i.e., a 150% increase to $8.75/kg) were examined (Figure 5). The results indicate that the supply chain can tolerate up to a 50% increase in black mass price ($5.3/kg) without any reduction in processing capacity but a 45% reduction in NPV ($10.1 billion). A further 50% increase in black mass price ($7/kg) would lead to 40% reduction in processing capacity and an 84% decrease in NPV ($2.9 billion).

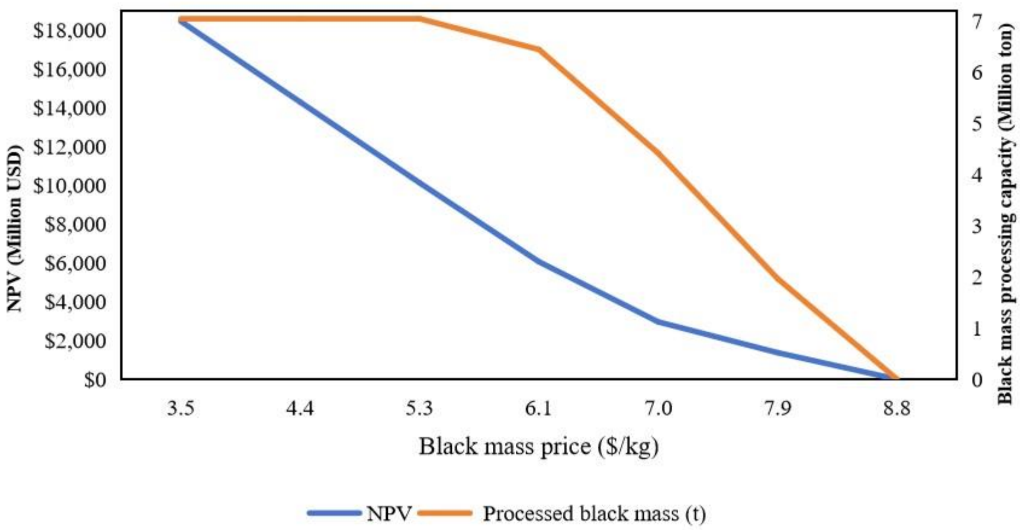

Figure 5. Change in net present value of implementing LIB bioleaching supply chain and its processing capacity in relation to the change LIB black mass purchase price.

5.2. Non-cooperative supply chain

The scenario analysis also considered the sustainability of the supply chain under non-cooperative situations. In particular, the analysis examined situations where the bioleaching of LIB is not vertically integrated. In this case, each upstream tier aims to maximize its own profit by selling its product to the next tier with a markup price, rather than cooperating to maximize the NPV of the entire supply chain. To evaluate this situation, a weighted matrix method was used to combine each tier's objective function, with the goal of minimizing the difference between the NPV of each tier and its ideal NPV*. Here, the ideal NPV* for each tier was obtained by solving the original model (Eqs 7–25) shown in section 3.2 with key assumptions shown in Table 2 for maximizing the profit of the tier (i.e., revenue from selling the tier's product—the tier's setup/operating/transportation costs). Then, the MILP model described in Section 3.2 was solved with the new objective function (Eq 26).

$$Min\ d = |NPV_{sort} - NPV^{*}_{sort}| + |NPV_{acid} - NPV^{*}_{acid}| + |NPV_{bioleaching} - NPV^{*}_{bioleaching}| \quad (26)$$

Figure 6 shows the resulting NPV of the entire supply chain under different scenarios, considering changes in the price of non-recyclable paper and acid from their base case ($0.089/kg and $1.3/kg, respectively). Green cells show the scenarios under which the supply chain could operate profitably. As the color shifts towards red, the profitability of one or more tiers decreases, resulting in an overall decrease in the supply chain's economic sustainability. The red cells indicate scenarios where the supply chain could not sustain as one of the tiers lost profitability. For example, increasing non-recyclable paper price by 100% while keeping the price of acid the same would jeopardize the profitability of acid producers, leading to an unsustainable supply chain. The same trend is observed with increasing the acid price and its effect on the profitability of bioleaching facilities. Overall, the results suggest that the supply chain cannot tolerate any increase in non-recyclable paper price greater than 900% ($0.89/kg) or any increase in acid price greater than 550% ($8.5/kg) in any scenario.

Acid / Non-recyclable paper		Price markup					
		0%	100%	200%	300%	400%	500%
Price markup	0%	18	18	18	14	9	-
	100%	-	18	18	18	14	1
	200%	-	18	18	17	14	9
	300%	-	18	18	17	12	7
	400%	-	18	18	17	12	6
	500%	-	18	18	16	12	-

Figure 6. The entire supply chain NPV (in $B) over the planning horizon in different pricing scenarios.

6. Conclusions

The amount of waste lithium-ion batteries (LIBs) will increase dramatically in the near future due to a large number of LIBs reaching the end of their useful lives and manufacturing scraps from increasing LIB production. Environmental concerns about the disposal of waste LIBs and the opportunity in recovering critical materials makes LIB recycling inevitable. Bioleaching technology showed the economic and environmental potentials as an alternative for current recycling methods [22,34]. As an emerging technology for LIB recycling, bioleaching requires proper supply chain design to connect with the required upstream tiers, which is adapted to the technology and incorporates future trends in main factors affecting the optimal configuration of the network.

We evaluated the potential of adopting bioleaching technology to create a profitable supply chain for LIB recycling. Main influential factors on the economic performance of the bioleaching process of spent batteries in the literature were considered in this study by projecting the metal price and change in metal composition of the black mass. The net present value of the supply chain was estimated around $18.4 billion for operating over 10 years to achieve the maximum recycling capacity of 900,000 tons of LIB black mass (~1.8 million tons of spent LIBs) per year in the United States.

The economic viability of the technology was found to be particularly sensitive to the cost associated with purchasing black mass, which was responsible for more than 60% of the entire supply chain cost. The research assessed the sustainability of the supply chain in regard to price changes of black mass. The findings indicate that the supply chain demonstrated resilience in tolerating an increased black mass price of up to $5.3/kg, without experiencing any decrease in the processing capacity of wasted LIB. The prices higher than $8.7/kg of LIB black mass jeopardized the economic viability of the supply chain. Additionally, the non-cooperative situation in the bioleaching supply chain was examined where each tier tried to maximize its own profit. Scenarios under different price markups for non-recyclable paper and acid revealed how the overall profitability of the supply chain changes with regard to different pricing strategies of sortation facilities and acid producers. Maximum prices for non-recyclable paper and acid prices were estimated as $0.89/kg and $8.5/kg, respectively, over which the supply chain could no longer be sustainable in any pricing scenario.

While this study did not assess the environmental impact of adopting bioleaching technology, the findings provide valuable insights into the economic feasibility of the technology and its potential to create a sustainable supply chain for LIB recycling. Further studies that incorporate other sustainability aspects are needed to provide a comprehensive evaluation of the technology's sustainability. Overall, the findings of this study can be used to inform decision-making processes for stakeholders involved in lithium-ion battery recycling supply chains.

Use of AI tools declaration

The authors declare they have not used Artificial Intelligence (AI) tools in the creation of this article.

Conflict of interest

Hongyue Jin is editor-in-chief for Clean Technologies and Recycling and was not involved in the editorial review or the decision to publish this article. All authors declare that there are no competing interests.

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