

Comparison of Hydrometallurgical and Hybrid Recycling Processes for Lithium-ion Battery: An Environmental and Cost Analysis

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Abstract

There is a strong emphasis on shifting to Electric Vehicles (EVs) throughout the globe for reducing the impact on global warming. Lithium-ion Batteries (LIBs) are predominantly used in EVs and these can be a significant threat to the environment if not disposed safely. Two routes namely pyrometallurgical and hydrometallurgical processes have been proposed in the literature for recycling LIBs. In this paper, we focus on quantifying the environmental impact of hydrometallurgy and hybrid (pyrometallurgy followed by hydrometallurgy) recycling processes and determine the option which has a lower impact on the environment. Life Cycle Assessment (LCA) is used for the environmental impact assessment of LIB recycling. The analysis is performed assuming ideal conditions for the hydrometallurgical and hybrid process in the inventory analysis. The reason being it was possible to determine the reaction yields only by theoretical calculations and not by field or experimental study. We have provided the lower bounds of impact by assuming a 100% conversion rate for the calculation of reaction yields in inventory analysis. Hence, this paper serves as a precursor or feasibility study for the policy-making step before setting up the battery recycling plants. The main contribution of this research is stepwise methodology development for LCA of LIB recycling and theoretical estimation of the reaction yields for inventory analysis. It was found that the cathode, anode, and foil of the LIB contribute most to the impact compared to the rest of the inputs. The impact due to separator, cell case, cables, leaching agent, slag-forming agent and reducing agent were minimum. The results show that the environmental impact of the hybrid process is found to be slightly better than the hydrometallurgy process across all environmental indicators. Further, a simple cost analysis was performed, and it also supports the hybrid process as a better option due to lesser variable costs.

Introduction

Globally, there is a gradual shift in the automotive industry towards Electric Vehicles (EVs) to reduce the impact on global warming (Frischknecht and Flury 2011). Lithium-ion Batteries (LIBs) are predominantly used in EVs because of their high energy density, low self-discharge, low maintenance cost and no requirement for priming (Gaines 2014). The disposal of the LIBs has become a growing concern since a significant amount of waste is generated every year (Miller 2015). There is an urgent need to dispose LIBs in an environmentally friendly manner. There is also an emphasis on recycling batteries since lithium is not available in abundance (Pehlken et al. 2015). Therefore, it is essential to improve the waste LIB collection rate to establish a sustainable battery recycling industry in the future (Wang and Yu 2020). Studies have shown that the current recycling processes based on pyrolysis and cryogenic methods for recycling LIBs are not energy efficient and cause more pollution to the environment due to the release of toxic by-products (Swain 2017). Also, it is not economical to recover the lithium from the batteries since it occurs in the form of compounds like LiMO_2 (Guo et al. 2017). Besides, aluminium, graphite, copper, organic solvent, lithium hexafluorophosphate, polyethylene or polypropylene are coated on the metal foil in the form of powder and needs to be extracted out separately during recycling (Zeng et al. 2014).

There are many processes for recycling LIBs, the two most common processes that are widely used are pyrometallurgical and hydrometallurgical recycling processes (Sonoc et al. 2015; Broussely and Archdale 2004). Another main recycling process used for LIB recycling is the direct recycling process.

Pyrometallurgical, Hydrometallurgical and Direct Recycling Process

An integral part of the recycling process is the pre-treatment step which is common to both pyrometallurgical and hydrometallurgical processes without which recycling is not possible (Reuter et al. 2003). In the pre-treatment step, sorting and exposing of battery cells are performed to segregate plastic, electronic components and other parts. This is followed by the vacuum thermal treatment at a very low temperature using nitrogen through cryogenic methods to deactivate the cells (to prevent explosion) and separate the electrolyte condensate (Boyden et al. 2016). The deactivated cells are mechanically processed to sort out iron-nickel, aluminium and electrode foil fraction by crushing and material separation. The electrode foil fractions are pelletized by adding binder and slag-forming components and are then fed to a high-temperature furnace to recover metals at around 1500°C (Trager et al. 2015). Limestone and coke are added as slag-forming and reducing agents respectively to the furnace for carbo-reductive smelting to reduce the metals as alloys of cobalt, nickel, copper and iron (Vieceli et al. 2018). This is the pyrometallurgical recycling process, the remaining metals like lithium, aluminium and manganese cannot be recycled using this process. Therefore, it comes out as slag which further needs to be processed by hydrometallurgical steps (Wang and Friedrich 2015). Figure 1 shows a simple block diagram for the pyrometallurgy recycling processes.

In hydrometallurgy, leaching is performed at temperatures 50–80°C to extract out the metals into the solution using suitable reagents like sulphuric acid, hydrogen peroxide. The dissolved metals are recovered from the solution through precipitation using reagents like sodium hydroxide, potassium carbonate, sodium sulphide and finally refined (Ferreira et al. 2009; Shin et al. 2005). Figure 2 shows a simple block diagram for the hydrometallurgy recycling processes.

As per the literature, pyrometallurgy and hydrometallurgy recycling are the two most commonly used recycling processes for Li-ion battery recycling. Another method that is used for recycling batteries is the direct recycling process. The direct recycling process also involves shredding or dismantling of the cells to recover aluminium and copper foils as metals. These metals could be further separated from each other for reuse to manufacture new cathode material. Similar to the hydrometallurgy process, it doesn't need much energy and can be carried out at room temperatures. It retains the crystal structure of the cathode, unlike the hydrometallurgy process where it is dissolved into the aqueous solutions due to the use of strong acids. Hence cost-wise, direct recycling is cheaper since it doesn't involve the use of strong acids and could also be used locally for home scrap. One limitation of direct recycling is that the output will be a mixture with significantly less value due to impurities and must be segregated based on the type of cathode post recycling (Gaines 2018).

Life-cycle Assessment (LCA)

Life-cycle Assessment (LCA) is used to quantify the environmental impact of a product or process throughout its life cycle starting from raw material extraction, production, use and End-of-Life (EoL) (ISO 14040 2006). Generally, the environmental impact of the EoL stage is between one to four orders of magnitude lower compared to all other stages of the total life cycle (Kirkpatrick et al. 2000). The recovery yield of battery materials was also found to affect the environmental impact. Future developments must be in the direction of recovering non-metallic materials to improve the recovery yield (Li et al. 2016). The environmental benefits are relatively larger than the environmental burdens associated with the dismantling and recycling processes (Jeong et al. 2007). Recycling ferrous metals and shredding processes are critical to improving the recycling rate to make the recycling treatment environmentally sound (Funazaki et al. 2003). In another study, it was found that the energy consumption in the use stage accounts for maximum consumption and remanufacturing possesses great economic value and practicability. Remanufacturing can potentially reduce the environmental impact due to less energy and resource consumption compared to manufacturing of the new product (Ming and Ming 2005). In a quantitative analysis study of carbon footprints and environmental impact assessment for lithium-ion secondary batteries, it was found that the carbon dioxide equivalence for the raw materials assembly, production and transport stage was lowest for the lithium-ion secondary battery followed by solar cell and nickel-metal hydride battery (Liang et al. 2017). The reason for lower GHG emission is due to the small volume and lightweight of the LIBs. Also, the manufacturing of LIBs is an energy-intensive process due to electricity production (Tian et al. 2017). The impact of emissions in the production phase is the highest compared to other phases and recycling could potentially reduce this impact by 10%-30% (Zackrisson et al. 2016). Water is preferred as a solvent for casting cathode and anode of LIBs and improvements in battery technology have led to a decrease in the environmental impact of the production phase almost to the level of use phase of LIBs (Zackrisson et al. 2010). The environmental impact due to losses in internal battery efficiency in fossil fuel-based vehicles is two to six times more than the losses due to LIB weight in electric vehicles. So, we must switch to alternatives that cause lesser impact due to lesser loss in battery efficiency (Gaines et al. 2011). It has been reported that LIBs cause a significant impact on the environment throughout their life cycle right from raw material extraction, production, use to disposal (Hao et al. 2017). This will lead to more environmental impact in the future and defeats the whole purpose of replacing conventional fossil fuel-based vehicles with electric vehicles. Therefore, a need has been realised to quantify the environmental impact of the LIB life cycle and identify ways to mitigate the impact (Peters et al. 2017). Additionally, improving fuel economy is identified as a crucial factor in maximizing the emission reduction potential in large-scale applications of electric vehicles using LIBs (Xiong et al. 2021).

In this study, the content analysis method has been used for the literature review. It was found that no study has been made so far in the literature that quantifies the environmental impact of the Li-ion battery recycling process. In this paper, we have compared and quantified the two most commonly used recycling processes for Li-ion battery recycling in terms of both environmental impact and cost to determine the

better option. Before doing an actual experimental study by the recycling practitioners, we are studying the feasibility of this process through a theoretical study for the policy-making step of whether recycling will have an impact on the environment. This study can be a further input to the policymakers for cost-benefit analysis and determining the inflexion point. This has not been applied in battery recycling so far in the literature and hence we are estimating the environmental and cost impact as a preliminary or feasibility study. In this paper, we have restricted the scope by considering only the recycling phase of the disposal of the batteries to quantify the environmental impact. Two recycling processes have been considered namely hydrometallurgical and hybrid (pyrometallurgy followed by hydrometallurgy) recycling processes. Since the pyrometallurgical process cannot recover metals like lithium, manganese and aluminium, it is generally followed by the hydrometallurgical process to recover these metals. Therefore, we compare the environmental impact of hydrometallurgy and the hybrid process. We have used LCA as a tool to quantify the environmental impact of the recycling processes to determine the option which has a lower impact on the environment. The mass of the inputs is determined from the stoichiometry of chemical reactions and assuming a 100% conversion rate. But in reality, the impact will be more than what we estimate and so in this paper, we are only providing a lower bound estimate of the environmental impact. Here, the LCA is cradle to grave and since we are comparing the environmental impact between the two routes of recycling, the difference is going to be only on the recycling process (i.e., the impact starting from mining, manufacturing and use phases will be the same). Further, a simple cost analysis is also performed to determine the better option out of the two processes.

Methods

In this paper, the LCA method is used to quantify the environmental impact of the LIB recycling processes. As per the ISO 14040 guidelines, there are three stages in LCA namely Goal and Scope Definition, Inventory Analysis and Impact Assessment as explained in the following sections. Lastly, we have performed a simple cost analysis by comparing the fixed and variable cost components of the recycling processes.

Goal and Scope Definition

The goal of this study is to quantify the environmental impact of the hydrometallurgical and hybrid recycling processes for the LIBs and determine the greener option out of the two processes under study. The findings of this study can be useful to the end-of-life vehicle handling practitioners who intend to set up a recycling plant for the LIBs.

The functional unit for the study is one tonne of EoL LIBs. The functions of the product system in scope are pyrometallurgical and hydrometallurgical recycling processes. The reference flow is determined by the mass of the materials present in the LIB. In this study, we define the boundary by considering only the recycling phase in the EoL (disposal) stage of the battery life cycle. Figure 3 shows a typical battery treatment system with different phases.

The battery is collected and around 40% of the weight is recovered from dismantling that includes disassembly of components like cathode, anode, casing, frame, cables etc. Fluids like electrolytes are

also drained separately. This is followed by a recovery which includes reuse and remanufacturing. The parts that are obtained from disassembly can be either reused or remanufactured depending on their age and state of repair. Post this, the rest of the battery is shredded and separated for recycling. Magnetic and eddy current separation is used to remove ferrous and non-ferrous materials respectively. Further manual sorting is performed to segregate Automotive Shredder Residue (ASR) fractions that include plastic, glass, rubber, fibre, foam and dirt. The remaining light and heavy ASR fractions are incinerated for energy recovery where it is burned in an incinerator at a high temperature to recover energy in the form of heat. Finally, the unusable waste from recycling, reuse, remanufacturing and energy recovery operations go through an audit to target for zero landfill assessment and future material substitution before landfill.

Inventory Analysis

Composition of Lithium-ion Battery

The main input for the LCA is the composition of the battery. Since LCA is a cradle to grave approach, we analyse the environmental impact of the battery composition throughout its life cycle that includes mining, manufacturing, use and disposal. Though we consider the recycling phase of the disposal of batteries, the impact of the constituents involved in recycling is considered throughout the entire life cycle. For example, the impact of the anode (graphite) in recycling is considered starting from mining till it is disposed. Hence it is a cradle to grave approach though the LCA is performed for one process (recycling). In this paper, our scope is restricted to the recycling process in the disposal phase of the battery. Also, we are doing a comparison between two kinds of the recycling process. Therefore, the impact from the mining till use phase will be the same and the difference in impact will be observed only in the recycling.

LIB is composed of various components made of metals and non-metals. Table 1 presents the material composition and the mass of the constituents present in different components of the battery.

Hydrometallurgical Recycling Process

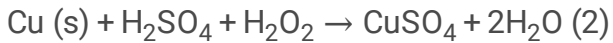
Leaching is the process of extracting substances from a solid by dissolving them in a liquid, either naturally or through an industrial process. Here, H_2SO_4 (sulphuric acid) and H_2O_2 (hydrogen peroxide) are used as leaching agents. Lithium cobalt oxide (LiCoO_2) reacts with H_2SO_4 and H_2O_2 to form Li_2SO_4 (lithium sulphate), CoSO_4 (cobalt sulphate), H_2O (water) and oxygen (O_2). The reaction is,



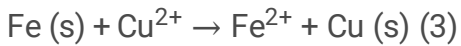
From the stoichiometry, we determine that 9.08 kg of LiCoO_2 will require 13.64 kg of H_2SO_4 and 1.58 kg of H_2O_2 . The mass of Li_2SO_4 and CoSO_4 produced will be 5.1 kg and 14.37 kg respectively.

Similarly, the reactions for LiNiO_2 (lithium nickel oxide) and LiMnO_2 (lithium manganese oxide) are consistent with reaction (1). Here, NiSO_4 (nickel sulphate) and MnSO_4 (manganese sulphate) are produced instead of CoSO_4 and the rest of the products are the same as (1).

Cu (copper) reacts with H_2SO_4 and H_2O_2 to form CuSO_4 (copper sulphate) and H_2O ,



Copper cementation reaction is governed by the following reaction,



In the above Fe (iron) exists as iron powder and the mass of Cu present in the battery is 11.14 kg. Based on the stoichiometry, we determine the mass of H_2SO_4 , H_2O_2 required and CuSO_4 produced to be 17.18 kg, 5.96 kg and 27.97 kg respectively.

Precipitation is the creation of a solid phase when the concentration of a salt exceeds its solubility in a solution. CoSO_4 obtained in reaction (1) is precipitated using NaOH (sodium hydroxide) and the reaction is,



From reaction (1), we find that the mass of CoSO_4 is 14.37 kg and from stoichiometry, we determine the mass of NaOH required is 7.42 kg. The mass of Co(OH)_2 (cobalt hydroxide) and Na_2SO_4 (sodium sulphate) produced will be 8.62 kg and 13.17 kg respectively. Similarly, NiSO_4 and MnSO_4 are precipitated using K_2CO_3 (potassium carbonate) and Na_2S (sodium sulphide) respectively.

The precipitation reactions of $\text{Fe}_2(\text{SO}_4)_3$ (ferric sulphate) and $\text{Al}(\text{NO}_3)_3$ (aluminium nitrate) in the presence of water and sodium hydroxide is,



From the stoichiometry, we determine the mass of H_2O , FeO.OH (ferric oxy-hydroxide) and H_2SO_4 to be 17.46 kg, 43.1 kg and 71.31 kg respectively.



From the stoichiometry, we determine the mass of NaOH, Al(OH)_3 (aluminium hydroxide) and NaNO_3 (sodium nitrate) to be 37.52 kg, 24.39 kg and 79.73 kg respectively.

Lithium sulphate obtained during the leaching of LiCoO_2 , LiNiO_2 and LiMnO_2 is further precipitated using K_2CO_3 and the reaction is,



The mass of Li_2SO_4 is found to be 15.26 kg and based on the stoichiometry, we determine the mass of K_2CO_3 , Li_2CO_3 (lithium carbonate) and K_2SO_4 (potassium sulphate) to be 19.16 kg, 10.21 kg and 24.03 kg

respectively.

Table 1 below presents the complete life cycle inventory data required for the hydrometallurgical recycling process. The data includes the battery composition, amount of electricity and process water.

In Table 1, the mass is listed for the composition of one battery. However, for the LCA, we have quantified the impact considering one tonne of battery materials.

Table 1
Inventory Data for Hydrometallurgical Recycling Process

Inputs			
Component	Composition	Amount	Unit
Cathode	Lithium	1.97	Kg
	Cobalt	5.5	Kg
	Nickel	5.5	Kg
	Manganese	5.07	Kg
	Oxygen	8.8	Kg
Anode	Graphite	19.74	Kg
Casing	Steel	28.06	Kg
Electrolyte	Ethylene carbonate	14.46	Kg
	Lithium hexafluorophosphate	1.61	Kg
Cables	Copper	1.83	Kg
	Aluminium	0.197	Kg
	Steel	1.001	Kg
Frame	Steel	1.001	Kg
	Plastic	15.087	Kg
Foil	Aluminium	5.499	Kg
	Copper	9.306	Kg
Cell case	Aluminium	2.961	Kg
Separator	Polyethene	7.6	Kg
Leaching agent	Hydrogen peroxide	10.69	Kg
	Sulphuric acid	58.03	Kg
Precipitating agent	Sodium sulphide	7.19	Kg
	Sodium hydroxide	44.94	Kg
	Potassium carbonate	32	Kg
	Process water	17.46	Kg
Electricity		19.6	Kwh
Process water		0.1	Cubic metre

Inputs			
Outputs			
Product output	Cobalt hydroxide	8.62	Kg
	Nickel carbonate	11.04	Kg
	Manganese (II) sulphide	8.01	Kg
	Copper	11.14	Kg
	Iron (III) oxide-hydroxide	43.1	Kg
	Aluminium hydroxide	24.39	Kg
	Lithium carbonate	10.21	Kg
Emissions to water	Sodium sulphate	63.5	Kg
	Sulphuric acid	71.31	Kg
	Sodium nitrate	79.73	Kg

Hybrid Recycling Process

The Pyrometallurgical process can be used only for recycling cobalt (Co), nickel (Ni), copper (Cu), iron (Fe) and the remaining elements like lithium (Li), manganese (Mn) and aluminium (Al) coming out as slag which needs to be further processed by the hydrometallurgical process.

The reaction of LiCoO_2 (lithium cobalt oxide) with coke (C) as the reducing agent to form Li, Co and CO_2 (carbon-di-oxide) is as follows,



Similar reactions follow for LiNiO_2 . The reduction reactions of CuO (copper (II) oxide) and Fe_2O_3 (iron (III) oxide) are,



We determine the total mass of the reducing agent required to be 29.51 kg. We use the higher heating value of carbon which is 32.8 MJ/kg to obtain 967.93 MJ energy of coke. The mass of slag forming agent (limestone) is assumed to be the same as that of coke which is 29.51 kg.

The mass of the outputs in the chemical reactions was obtained by theoretical calculations since it was not possible to get the field or experimental data for the reaction yields. We have taken the typical composition of inputs from the literature and calculated the lower bounds of the environmental impacts

assuming ideal conditions (i.e. 100% conversion rate). However, in reality, the conversion rate will not be 100% and the impact will be higher due to loss factor or percentage. The paper aims to give an estimate of the lower bound impact to the practitioners if they intend to set up battery recycling plants. In reality, the same steps can be followed to determine the reaction yield by performing the experiments. The contribution of this paper is to perform a feasibility study through LCA inventory analysis and quantifying the environmental impact. The methodology development along with the theoretical contribution and stepwise LCA approach has been the main objective of this paper. Though it is a theoretical study, this research has been performed by consulting experts from chemical engineering and potential users of such batteries from automotive engineering.

Impact Assessment

In the impact assessment phase, the inventory data is used to calculate the environmental effects on emissions to air, water and land. Here impact on resource depletion, human health and solid wastes are quantified and obtained as output and the results are interpreted (ISO 14040 2006). CML-IA impact assessment method developed by the Centre of Environmental Science, Leiden University, Netherlands is used to quantify the environmental impact indicators. For this, we use the inventory data and the impact categories recommended by the International Reference Life Cycle Data System (ILCD) Handbook in this study. Eleven environment impact indicators are quantified namely, abiotic depletion (AD), abiotic depletion – fossil fuels (AD - FF), global warming potential (GWP), ozone layer depletion (OD), human toxicity (HT), freshwater aquatic eco-toxicity (FWAE), marine aquatic eco-toxicity (MAE), terrestrial eco-toxicity (TE), photochemical oxidation (PO), acidification (AC) and eutrophication (EU) (Wolf et al. 2010; Guinee 2002).

Cost Analysis

A simple cost analysis was performed to compare the hydrometallurgical and hybrid processes. Two kinds of costs are involved namely fixed and variable costs.

Fixed cost includes facility land cost, construction cost and cost of the equipments (mainly furnace and shredding equipment). The hybrid process would incur more fixed cost than the hydrometallurgical process since additionally a furnace is required for the pyrometallurgical process and not the hydrometallurgical process. All the other components of the fixed cost would be the same for both processes.

Variable cost includes the cost of reagents (leaching, precipitating, slag-forming and reducing agents) and electricity. The data for INR/unit for the reagents is taken from the India Mart (an Indian e-commerce company) website. Table 2 shows the estimation of different components of variable costs for recycling batteries. The inputs regarding the quantity per battery are taken from the inventory analysis and in turn multiplied by INR/unit to get the estimated cost per battery for both hydrometallurgy and hybrid processes. The average cost per battery of these two processes is taken to estimate the variable cost of recycling batteries.

Table 2
Variable Cost of Recycling Battery

Inputs	INR/unit	Quantity/battery - Hydro (kg)	Quantity/battery - Hybrid (kg)	Cost/battery – Hydro (INR)	Cost/battery – Hybrid (INR)
Hydrogen peroxide	32	11	2	342	50
Sulphuric acid	15	58	14	870	203
Sodium sulphide	30	7	7	216	216
Sodium hydroxide	54	45	37	2427	2026
Potassium carbonate	53	32	19	1696	1015
Coke	18	-	29		531
Limestone	3	-	29		88
Electricity	6	20	112	124	711
			Total cost	5675	4842

Results And Discussion

Table 3 and Fig. 4 present the percentage contribution on the environmental impact of different components in the hydrometallurgical recycling process across eleven indicators. Table 4 and Fig. 5 present the percentage contribution of the hybrid recycling process across eleven indicators.

Table 3

Percentage Environmental Impact due to Components across Eleven Indicators (Hydrometallurgical Recycling Process)

Impact category	AD	AD (FF)	GWP	OD	HT	FWAE	MAE	TE	PO	AC	EU
Cathode	8%	22%	27%	10%	9%	22%	15%	12%	58%	61%	14%
Anode	32%	13%	12%	32%	35%	33%	10%	31%	15%	15%	38%
Cables	6%	2%	2%	5%	6%	6%	2%	6%	2%	2%	6%
Casing	1%	4%	5%	1%	9%	2%	2%	15%	2%	1%	2%
Frame	0%	16%	15%	1%	1%	4%	2%	1%	2%	2%	3%
Electrolyte	2%	9%	7%	3%	6%	1%	47%	1%	2%	2%	1%
Foil	43%	9%	9%	27%	31%	29%	11%	28%	13%	13%	33%
Cell case	8%	1%	1%	0%	1%	1%	2%	1%	1%	0%	0%
Separator	0%	5%	2%	0%	0%	0%	0%	0%	0%	0%	0%
Leaching Agent	0%	4%	2%	2%	0%	0%	1%	0%	2%	2%	0%
Precipitating agent	0%	14%	17%	18%	2%	3%	7%	5%	3%	3%	2%
Electricity	0%	1%	2%	0%	0%	0%	1%	0%	0%	0%	0%

Table 4
Percentage Environmental Impact due to Components across Eleven Indicators (Hybrid Recycling Process)

Impact category	AD	AD (FF)	GWP	OD	HT	FWAE	MAE	TE	PO	AC	EU
Cathode	8%	21%	26%	10%	9%	22%	15%	12%	57%	61%	14%
Anode	32%	12%	11%	32%	35%	32%	10%	31%	15%	15%	37%
Cables	6%	2%	2%	5%	6%	6%	2%	6%	2%	2%	6%
Casing	1%	4%	5%	1%	9%	2%	2%	15%	2%	1%	2%
Frame	0%	15%	15%	1%	1%	4%	2%	1%	2%	2%	3%
Electrolyte	2%	9%	7%	3%	6%	1%	46%	1%	2%	2%	1%
Foil	43%	8%	9%	27%	30%	29%	11%	28%	13%	13%	33%
Cell case	8%	1%	1%	0%	1%	1%	2%	1%	1%	0%	0%
Slag forming agent	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%	0%
Reducing agent	0%	6%	3%	2%	2%	0%	0%	0%	2%	1%	1%
Separator	0%	4%	2%	0%	0%	0%	0%	0%	0%	0%	0%
Reagent	0%	11%	12%	15%	1%	2%	5%	4%	2%	2%	2%
Electricity	0%	7%	9%	2%	1%	1%	5%	1%	1%	1%	1%

To understand the impact on the environment of the two processes, we compare in Table 5 the contributions of the two processes across all the indicators.

Table 5
Comparison of Environmental Impact between Hydrometallurgical and Hybrid Process

Impact category	Unit	Hybrid Process	Hydrometallurgy Process	Percentage Difference
AD	kg Sb eq	0.43767	0.43763	-0.01%
AD – FF	MJ	84821.71	89677.35	5.72%
GWP	kg CO ₂ eq	6409.03	6752.73	5.36%
OD	kg CFC-11 eq	0.00159	0.00157	-1.78%
HT	kg 1,4-DB eq	12196.12	12489.38	2.40%
FWAE	kg 1,4-DB eq	249.22	250.11	0.36%
MAE	kg 1,4-DB eq	8001467.60	8201491.10	2.50%
TE	kg 1,4-DB eq	46.70	46.67	-0.08%
PO	kg C ₂ H ₄ eq	8.60	8.70	1.11%
AC	kg SO ₂ eq	208.33	207.91	-0.20%
EU	kg PO ₄ [—] eq	22.11	22.28	0.77%

It can be observed that cathode, anode and foil contribute the most across all the indicators for both hydrometallurgy and hybrid recycling processes. For both hydrometallurgy and hybrid process, cathode causes the maximum impact across AD-FF (22% and 21% respectively), GWP (27% and 26% respectively), PO (58% and 57% respectively), AC (61% and 61% respectively) whereas anode causes the maximum impact across OD (32% and 32% respectively), HT (35% and 35% respectively), FWAE (33% and 32% respectively), TE (31% and 31% respectively) and EU (38% and 37% respectively). This is attributed to the fact that these inputs have a high percentage of metals in them as compared to other inputs. The significant contribution to impact arises from mining, manufacturing and use. Similarly, the impact due to separator, cell case, cables, leaching agent, slag-forming agent and reducing agent are minimum as they have a low percentage composition of metals. Foil causes a maximum impact on AD (43%) in both hydrometallurgy and hybrid processes. Electrolyte causes a maximum impact on MAE in both processes (47% and 46% respectively). Since the pyrometallurgical recycling process uses a furnace, the impact due to electricity is more in the hybrid process than the hydrometallurgy process. There is a significant amount of impact due to frame in AD-FF (16% and 15% respectively) and GWP (15% and 15% respectively) for hydrometallurgy and hybrid processes. Casing causes a significant impact on TE (15% and 15% respectively) for hydrometallurgy and hybrid processes. The precipitating agent causes a significant impact on AD-FF, GWP and OD. The percentage contribution is 14%, 17% and 18% respectively for the hydrometallurgy process. The contribution of reagent on AD-FF, GWP and OD is 11%, 12% and 15%

respectively for the hybrid process. From Table 5, we observe that the overall hybrid process causes a slightly lesser environmental impact compared to the hydrometallurgical process.

From a cost analysis perspective, we find that the variable cost of the hybrid process is less compared to the hydrometallurgical process. The variable cost per battery of hybrid process is ~ 833 INR less than hydrometallurgical process due to the more quantity of reagents required for leaching and precipitation process. Though the fixed cost of the hybrid process is more due to the extra cost of the furnace, we don't consider this as a deciding factor since it is a one-time investment and the variable cost would eventually even out in the long term as large numbers of batteries are recycled. Also, the hybrid process has a better recycling rate than the hydrometallurgy process especially for the elements like Li, Mn and Al. Therefore, we suggest the hybrid process as a better option based on the environmental and cost perspectives.

Conclusion

In this paper, we have used LCA to quantify and compare the environmental impact of the hydrometallurgy and hybrid recycling process. Since we are comparing the environmental impact between the two routes of recycling, the difference is going to be only on the recycling process. Hence, LCA is performed only on recycling. Before doing an experimental study by the recycling practitioners, we are studying the feasibility of this process through a theoretical study. This is for the policy-making step of whether recycling will have an impact on the environment since LCA has not been applied in battery recycling so far. We have considered one tonne of LIB as the basis functional unit and defined the boundary by considering only the recycling phase of the EoL disposal of the battery. We theoretically estimated the inventory data that includes the input data, product output, emissions to air, water for the LCA based on the stoichiometry and yield of the chemical reactions of the pyrometallurgical and hydrometallurgical recycling processes. The conversion rate was assumed to be 100% for the chemical reactions however in reality, there will be a loss factor or percentage. So, we have only provided an estimated lower bound for the environmental impact and the actual impact would be higher. Using the inventory data, we quantified the environment indicators using the CML-IA method for the impact assessment. We found from the results that the cathode, anode and foil of the LIB contribute most to the impact compared to the rest of the inputs. The impact due to separator, cell case, cables, leaching agent, slag-forming agent and reducing agent were minimum. We found that in terms of both environmental and cost aspects, the hybrid process is better compared to the hydrometallurgical process. Hence, we suggest that the hybrid process can be used by the practitioners to recycle LIBs on a large scale.

Declarations

Author Contributions: All the authors have contributed to the whole development of the manuscript: designing the research, performing the calculations, writing the text, discussing the results, and obtaining the conclusions.

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Conflicts of Interest: The authors declare no conflict of interest.

Data Availability Statement: The authors confirm that the data supporting the findings of this study are available within the article.

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Figures

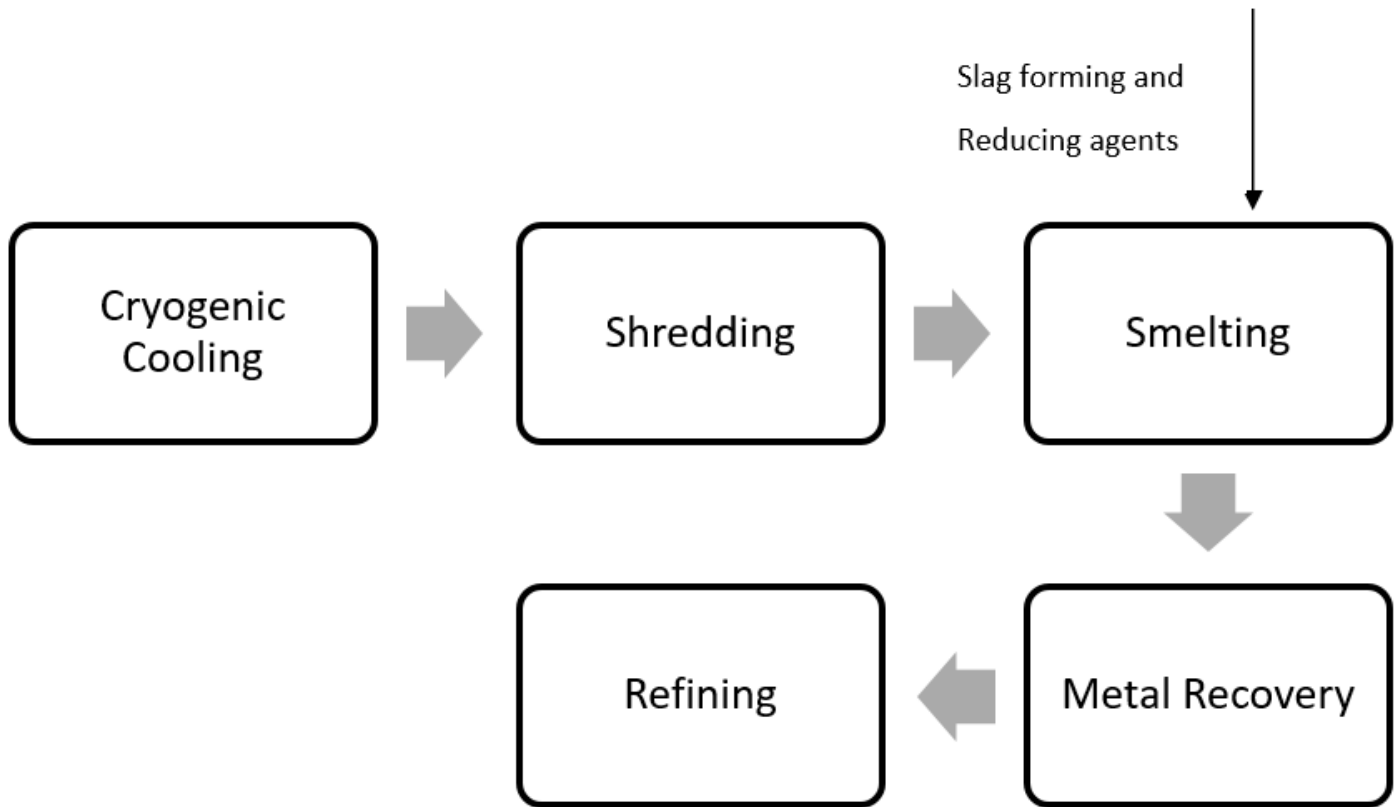


Figure 1

Pyrometallurgy Recycling Process

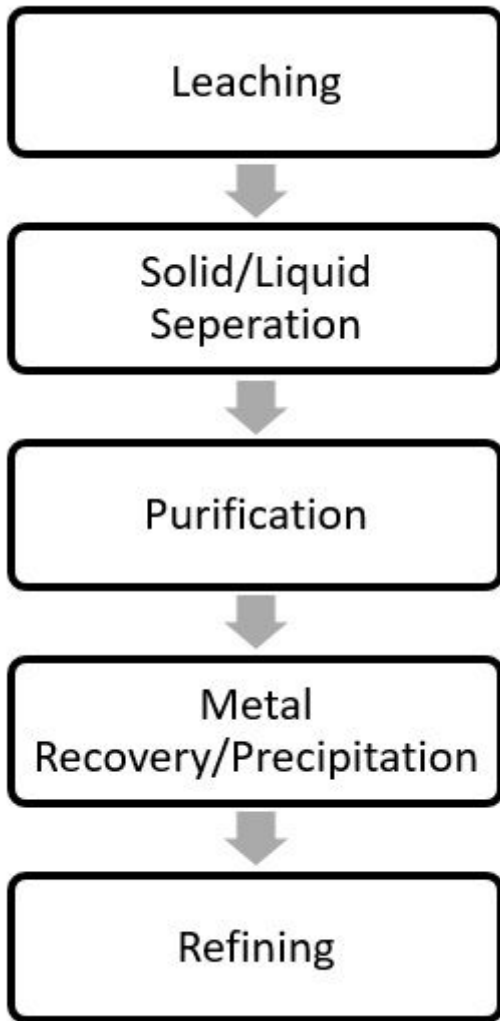


Figure 2

Hydrometallurgy Recycling Process

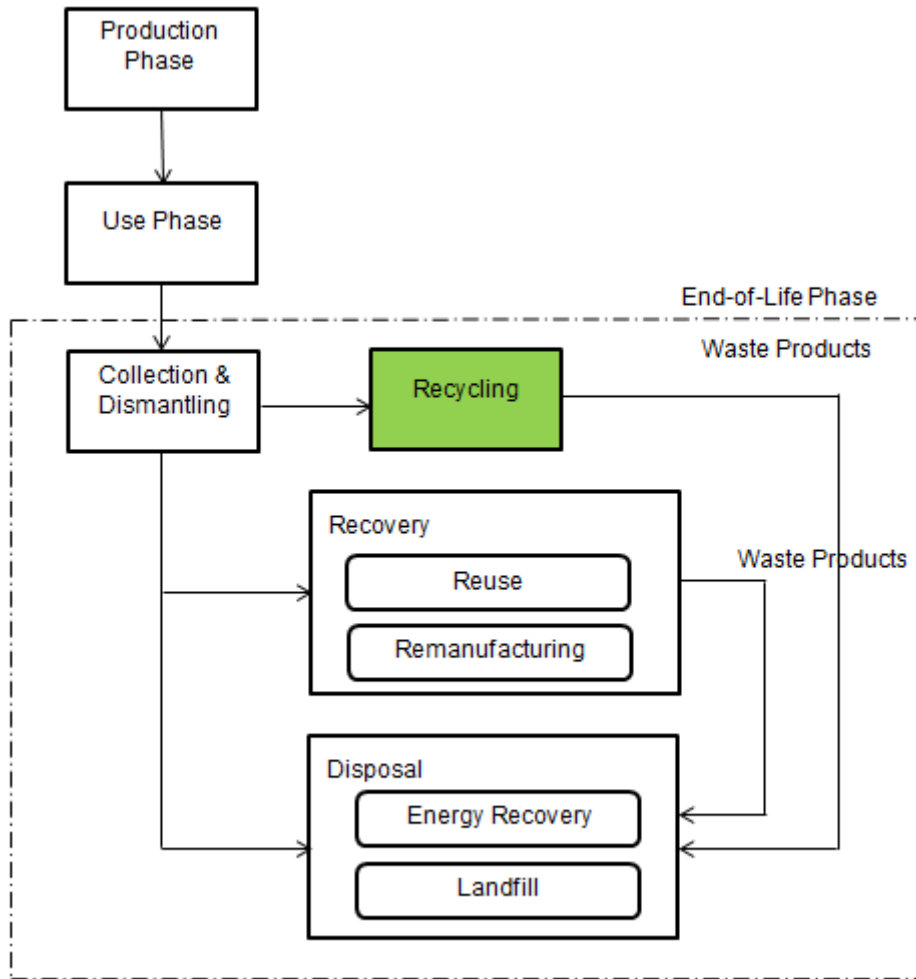


Figure 3

Battery Treatment Process

Hydrometallurgical Recycling Process

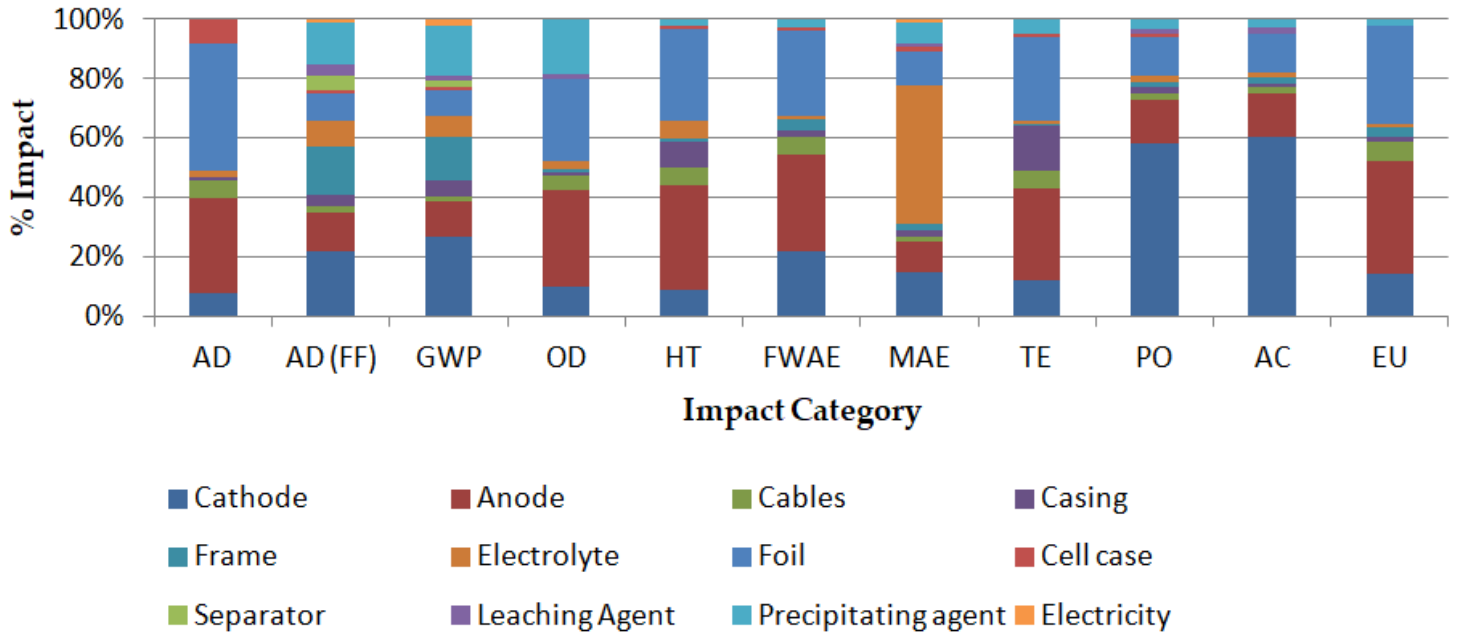


Figure 4

Graphical Representation of Percentage Impact due to Components across Eleven Indicators (Hydrometallurgical Recycling Process)

Hybrid Recycling Process

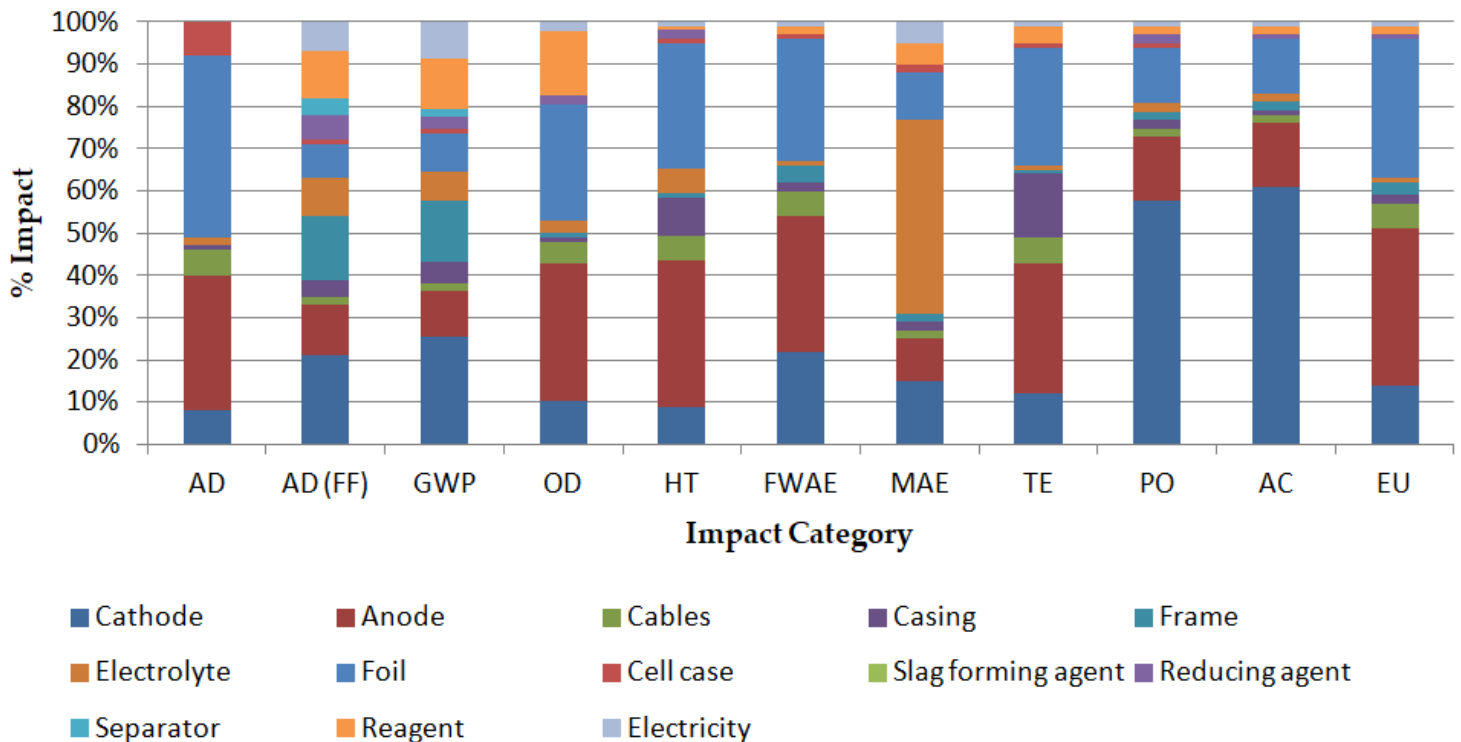


Figure 5

Graphical Representation of Percentage Impact due to Components across Eleven Indicators (Hybrid Recycling Process)

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