Update of Life Cycle Analysis of Lithium-ion Batteries in the GREET® Model

by

Q. Dai, J. Dunn, J. C. Kelly, and A. Elgowainy

Systems Assessment Group

Energy Systems Division

Argonne National Laboratory

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ACRONYMS

CATARC	China Automotive Technology and Research Center
CERC	Clean Energy Research Center
CVC	Clean Vehicle Consortium
LCA	life cycle analysis
LCI	life cycle inventory
LFP	lithium iron phosphate
LIB	lithium-ion battery
NMP	N-Methyl-2-Pyrrolidone
NMC	lithium nickel manganese cobalt oxide
ppmv	part per million by volume

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Qiang Dai, Jennifer Dunn, Jarod C. Kelly, and Amgad Elgowainy

Energy Systems Division
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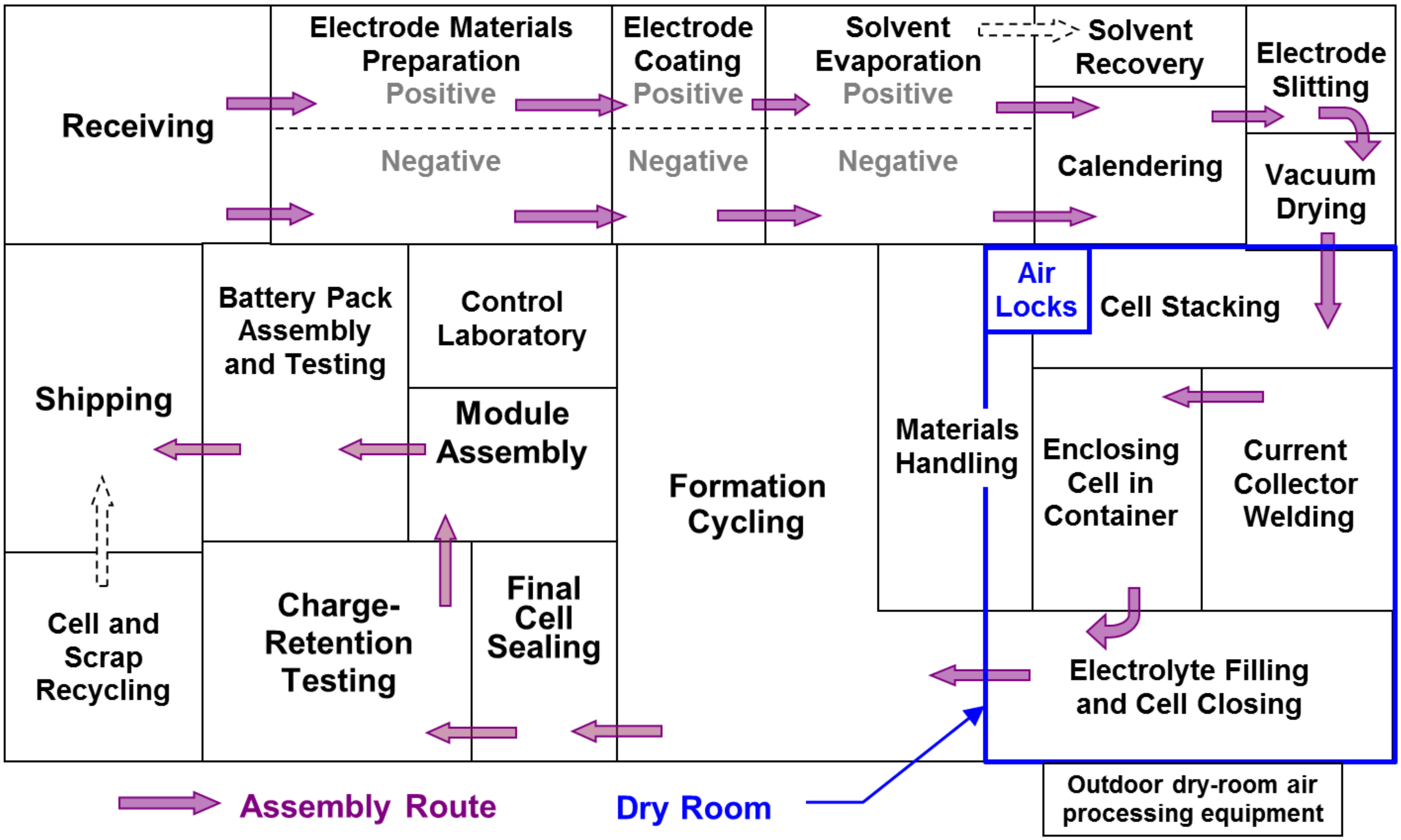
This memo documents updates for life cycle analysis of lithium-ion batteries (LIB) in the GREET® model. These updates were obtained through 1) our site visits to two LIB manufacturing facilities and one LIB recycling facility in China; 2) Argonne's latest modeling effort by Ahmed *et al* to support efficient simulation, analysis, and design of advanced LIB technologies. These updates therefore reflect the current status of lithium nickel manganese cobalt oxide (NMC) cathode material production and LIB manufacturing, and will be incorporated into GREET 2017.

1 LIB MANUFACTURING

Process energy demand for LIB manufacturing has been identified as an environmental hotspot in previous battery life cycle analysis (LCA) studies (Kim *et al* 2016, Dunn *et al* 2015a, and Ellingsen *et al* 2014). However, reported energy consumption for LIB manufacturing is based on engineering calculations (Wood III *et al* 2015, Dunn *et al* 2014) or pilot-scale battery manufacturing facilities (Ellingsen *et al* 2014), and therefore does not necessarily represent the actual energy consumption of the LIB industry. Kim *et al* are the first to report the environmental impacts of battery manufacturing at a commercial-scale facility. However, for proprietary reasons, life cycle inventory (LCI) data for LIB manufacturing and assembly is not disclosed (Kim *et al* 2016). Since greenhouse gas (GHG) and criteria air pollutant emissions are very sensitive to the assumed electricity mix and upstream processes of fuels used for LIB production, the results of Kim *et al*, which are based on national average electricity mix and natural gas processing in South Korea, may not be representative of LIB manufacturing elsewhere in the world.

LIB manufacturing is a complex process comprised of numerous sub-processes, as depicted in Figure 1. Note that solvents are needed for the electrode material preparation process, to make the electrode material a slurry for subsequent electrode coating. Currently, N-Methyl-2-Pyrrolidone (NMP) is typically used for the positive electrode (hereinafter referred to as cathode) materials, while water is typically used for the anode electrode (hereinafter referred to as anode) materials (Wood III *et al* 2015). Due to cost and environmental concerns, NMP is usually recovered after the solvent evaporation process and reused for future LIB production (Wood III *et al* 2015). Also note that some of the electrode production processes, as well as the entire cell assembly process, need to take place in a dry room, in which the moisture content of the air cannot exceed 100 parts per million by volume (ppmv), to prevent $LiPF_6$, the dominant electrolyte salt, from detrimental reaction with water (Ahmed *et al* 2016a).



The areas in this diagram for each processing step are approximately proportional to the estimated plant areas in the baseline plant.

Figure 1. Process Schematic for LIB Pack Manufacturing (Source: ANL 2017)

Due to its stringent requirement for moisture control, which is typically accompanied by rigorous temperature control, the dry room has been singled out as a significant contributor to the total energy requirement for LIB manufacturing in previous LIB LCA studies (Dunn *et al* 2015a, Ellingsen *et al* 2014). Meanwhile, Wood III *et al* contends that cathode drying and NMP recovery, together with cell wetting and formation, are the most energy-intensive processes for LIB manufacturing (Wood III *et al* 2015). Ahmed *et al* adopts a chemical process modeling approach, and finds that, for a full-scale LIB manufacturing plant with a production capacity of 100,000 automotive battery packs per year, cathode drying and NMP recovery, and dry room operation, are two determinants of the energy demand and cost of LIB manufacturing (Ahmed *et al* 2016a, Ahmed *et al* 2016b). While these studies improve the understanding of LIB manufacturing processes and their energy demand an LCI representative of commercial-scale LIB manufacturing is still lacking in literature to date.

As part of the collaborative effort of U.S.-China Clean Energy Research Center (CERC)'s Clean Vehicle Consortium (CVC), with the help of China Automotive Technology and Research Center (CATARC), the authors visited two LIB production facilities of two leading Chinese LIB manufacturers and one leading Chinese LIB recycler in March 2017. One LIB manufacturer provided detailed information on the energy and water consumption of their LIB manufacturing processes. The visited facility of this manufacturer on average operates 300 days per year, 20 hours per day, and produces 50,000 16Ah 3.2V lithium iron phosphate (LFP) cells together with 30,000 43Ah 3.7V NMC cells per day.

The collected energy consumption data is for NMC cells production. According to the engineer in charge of equipment, the NMC production line consumes electricity and steam. Electricity is primarily used to power 11 dehumidifiers and 4 industrial water chillers for process cooling, and the electricity consumption by the rest of the equipment is negligible. The 11 dehumidifiers run year-round. They have a collective rated power of 500kW, while the actual power is typically 300kW. The water chillers each has a rated power of 380kW, and the actual power ranges between 150 kW and 350 kW. Depending on the outdoor temperature, 1~4 chillers may run at the same time. Steam is exclusively used for electrode drying and dehumidification. Each process consumes 4~5 metric tons of steam per hour. Dry room operation (i.e. dehumidification and cooling) and electrode drying are therefore confirmed as the biggest contributors to energy consumption for LIB manufacturing. Water consumption for the entire facility is estimated to be 200~300 m^3 per day, of which LIB production (both LFP and NMC cells) accounts for 80%.

The consumed steam is from the municipal steam network of the city where the facility is located. Although municipal steam in that city is produced in a combined heat and power plant fueled by coal, to make the LCI more universally applicable, the steam requirement in metric tons is converted into heat requirement in MJ based on the temperature (250°C) of the municipal steam, assuming a boiler efficiency of 80%, which is the default in GREET. The estimated energy consumption for per kWh of cell produced, together with literature values normalized per kWh battery, is summarized in Table 1.

Table 1. Energy demand for LIB manufacturing (MJ/kWh battery produced)

	Ellingsen 2014	Wood 2015	Kim 2016	Ahmed 2016[4]	This study
Plant capacity (per year)	Pilot plant[1]	<1000 packs	1M cells (0.06 GWh)	100,000 packs (1 GWh)	12M cells[2] (2 GWh)
Cell information	20Ah 3.65V	52Ah 3.5V	15Ah 3.7V	N/A	43Ah 3.7V
Pack information	26.6 kWh	N/A	24 kWh	10 kWh	N/A
Total energy demand (MJ/kWh)	586~2318	1941	990[3]	175	119~168
Total energy demand by source (MJ/kWh)					
Electricity demand	586~2318	1941	340[3]	59	7~26
Heat demand	---	---	650[3]	116	112~142
Total energy by use (MJ/kWh)					
Dry room	N/A	N/A	N/A	10 (2 NG, 8 elec.)[a]	63~96 (7~26 elec., 56~71 steam)
Electrode drying (and NMP recovery)	N/A	1129	N/A	152 (114 NG, 38 elec.)[b]	56~71 (steam)
Cell formation and cycling	N/A	812	N/A	13 (Elec.)[c]	N/A
Pack assembly	0.01	N/A	10[3] (Elec.)	N/A	N/A
Data source	Battery manufacturer (Miljøbil Grenland)	Model	Battery manufacturer (LG Chem)	Model	Battery manufacturing facility visit

1. The capacity of the Miljøbil Grenland plant was not disclosed in the paper. However, Electrovaya, which acquired Miljøbil Grenland in 2012, reported an annual revenue of $2.8 million US dollars for 2013 (Electrovaya 2013, Electrovaya 2012). The Miljøbil Grenland plant is therefore estimated to be pilot-scale.
2. Estimated based on a rated capacity of 40,000 cells per day. The plant operates 300 days per year.
3. Estimated based on reported primary energy consumptions and GHG emissions, with GREET GHG emission factors. NG is short for natural gas. Elec. is short for electricity.
4. Includes Ahmed *et al* 2016a, Ahmed *et al* 2016b, and Ahmed *et al* 2016c. Superscripts a, b, and c represent separate Ahmed publications.

Specific energy consumption was not available for cell formation and charging, nor for pack assembly from the site visits. For cell formation and cycling, although the battery manufacturers were not willing to disclose the temperature and duration for their cell formation process, they provided us the number of charge-discharge cycles they used. One manufacturer uses 1.5 cycles (charge-discharge-charge), and the other uses 2.5 cycles. Both manufacturers claim that they reuse electricity from discharge, which makes sense considering their scale of production, rated at 2 GWh/year for both of them. Therefore, the energy consumption for cell formation and cycling can be estimated as the amount of electricity needed to charge the battery once, plus the amount of electricity to make up for discharge loss. Assuming a charging efficiency of 90%, and a discharge loss of 10%, the energy consumption for cell formation and cycling is estimated to be 1.2kWh electricity/kWh cell produced. For pack assembly, we noticed during our visits that it was done manually. Even if the process is automated in the future, to our knowledge, there are no energy-intensive steps, such as cooling or drying, involved in pack assembly. Therefore, it can be assumed that the energy consumption for pack assembly is negligible compared with that for cell production. In other words, the same energy consumption will be assumed for 1 kWh of battery cell produced and 1kWh of battery pack produced.

In conclusion, energy consumption for LIB manufacturing is estimated to be 170MJ/kWh battery produced, of which 30 MJ is electricity, and the remaining 140 MJ is heat, assumed from natural gas. Water consumption for LIB manufacturing is estimated to be 8.6 gallon/kWh battery produced. Again, water consumption for pack assembly is assumed to be negligible. It should be noted that in previous versions of our GREET model, environmental impacts for LIB are calculated using one process energy intensity on a per mass basis (mmbtu/ton battery produced) for all available LIB chemistries. Since cell production accounts for all of the process energy demand, while pack configuration can substantially affect the specific energy of a battery pack, which can skew the process energy intensity on a per mass basis, starting from GREET2017, process energy intensity on a per kWh basis will be used for LIB environmental impacts calculations. Although the data we collected represent NMC battery production, and we understand that for a different cathode chemistry or even a different NMC cell configuration, the electrode area could be different, and therefore the energy demand for electrode drying, we contend that the process energy intensity on a per kWh basis will not change significantly across different LIB chemistries, since energy use of the dry room depends on its size, regardless of its throughput (Ahmed *et al* 2016a). In the absence of commercial-scale LIB production process energy demand data for other chemistries, the same process energy intensity on a per kWh basis will be used for all available LIB chemistries in GREET. Changes to be incorporated into GREET 2017 are summarized in Table 2.

Table 2. Changes to LIB manufacturing LCI in GREET

	GREET 2016	GREET 2017
Process energy consumption	0.450 mmbtu/ton battery*	0.161 mmbtu/kWh battery
Process energy share	49% NG, 51% electricity	82.4% NG, 17.6% electricity
Process water consumption	N/A	8.6 gallon/kWh battery

*Equivalent to 0.0025~0.006 mmbtu/kWh battery available in GREET, depending on battery pack specific energy.

It should be pointed out that process energy intensity decreases with increasing production capacity, as shown in Table 1, probably due to economy-of-scale and more efficient process design. With a few Gigafactories (e.g., Tesla, Northvolt, each has a capacity over 30 GWh/year) planned worldwide, the process energy demand for LIB manufacturing may decrease in the future. It should be also pointed out that the electrode drying process is energy-intensive because of the use of NMP as the solvent for cathode slurry preparation. Due to NMP's low flammability limit in air, the concentration of NMP vapor needs to be carefully controlled during the drying process, which requires massive amounts of heated air (Ahmed *et al* 2016b). Electrode processing technologies using water-based solvents are being developed for LIB production, as water is cheaper, and doesn't pose environmental and health hazard (Wood III *et al* 2015). Should water replace NMP as the solvent for cathode materials, the energy requirement for electrode drying will be reduced significantly. These issues should be examined in future GREET updates.

2 NMC CATHODE MATERIAL PRODUCTION

In addition to process energy consumption for LIB manufacturing, active cathode materials, especially those containing nickel and cobalt, are also a predominant contributor to the lifecycle environmental impacts of LIB (Kim *et al* 2016, Dunn *et al* 2015a, and Ellingsen *et al* 2014). Most of the impacts can be traced back to the upstream mining, smelting, and refining processes of the metals. However, the synthesis process for the active material is also important. Existing LCIs for cathode synthesis processes in GREET2016 were derived from engineering calculations based on synthesis conditions reported in literature (Dunn *et al* 2015b). To leverage Argonne's LIB modeling expertise, we will update the energy consumption for NMC cathode material production, based on the chemical process model developed by Ahmed *et al*. The existing LCI for NMC in GREET2016 represents the production process for $LiNi_{0.4}Mn_{0.2}Co_{0.4}O_2$, a.k.a. NMC(424). As NMC has several variants, we will add production LCIs for $LiNi_{1/3}Mn_{1/3}Co_{1/3}O_2$ (NMC(111)), $LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$ (NMC(622)), and $LiNi_{0.8}Mn_{0.1}Co_{0.1}O_2$ (NMC(811)) to GREET2017. Since automotive LIB recycling is being commercialized worldwide, we will also add to GREET2017 a LCI for NMC(111) produced from hydrometallurgical recycling, based on information we collected from our site visit to a leading LIB recycler in China.

There are two primary pathways for NMC synthesis via co-precipitation: carbonate and hydroxide (Ahmed *et al* 2017). Ahmed *et al* modeled the carbonate pathway for NMC(111), while we use the hydroxide pathway in GREET. Therefore, the material consumption reported by Ahmed *et al* is not directly applicable to NMC production in GREET. However, the overall process flow for the two synthesis pathways are similar, and the same nickel, manganese, and cobalt starting materials (sulfates) are used in both pathways. Therefore, an 85% material yield for nickel, manganese, and cobalt, which is the lower end of reported Ni/Mn/Co yields by Ahmed *et al*, is applied to existing Ni/Mn/Co salts consumption for NMC synthesis in GREET, which was based on stoichiometry. In addition, their reported process water consumption, 26,200 gallons per day (4 gallon/kg NMC produced), is added to GREET, since water consumption for the carbonate pathway and the hydroxide pathway can be similar (Ahmed *et al* 2017). Furthermore, the lithium salt used in GREET will be changed from lithium hydroxide to lithium carbonate, because both Ahmed *et al* and the battery recycler reported consistent lithium carbonate consumption, which suggests better data quality.

For process energy consumption, Ahmed *et al* stated that switching to the hydroxide pathway would only result in small changes to the energy consumption of the process. Their energy demand estimates, 1019kW electric power load and 33kW thermal load for a plant producing 6,500 kg of NMC(111) per day, will therefore be incorporated into GREET (Ahmed *et al* 2017). The NMC synthesis process consists of two stages: 1) mixing nickel, cobalt, and manganese sulfates to produce the precursor, an Ni-Mn-Co carbonate or hydroxide co-precipitates, depending on the synthesis pathway; 2) mixing the dried precursor with lithium carbonate or lithium hydroxide, and then calcine the mixture to produce NMC. Ahmed *et al* identified the calcining kiln to be the largest contributor to the energy consumption at the plant, accounting for 800kW of the total electric load (Ahmed *et al* 2017). It is therefore estimated that the precursor production stage consumes 0.81kWh electricity and 0.12kWh heat (provided by natural gas, assuming an 80% boiler efficiency) per kg of NMC produced, and the calcination stage consumes 2.95kWh electricity per kg of NMC produced.

For other NMC variants, material consumptions are estimated based on stoichiometry, adjusted by the 85% material yield for Ni/Mn/Co salts. Since our preliminary calculations showed that changes in Ni/Mn/Co composition in NMC would not lead to significant changes in process energy demand or water use, the same energy consumption will be used for all NMC variants, so is the same water consumption.

The Chinese battery recycler we visited recovers Ni, Mn, and Co from spent LIB batteries, and produces NMC precursor from these recovered materials. For the produced NMC precursor, they either supply it to other battery material manufacturers, or ship it to another facility of theirs to produce NMC cathode material. The overall process for NMC production from hydrometallurgical recycling of spent LIB is depicted in Figure 2. No specific material and energy consumption data were provided by the recycler during our site visit. However, they referred us to a paper they published, which contains LCI information for their process (Xie *et al* 2015). In their paper, they point out that calcination is the most energy-intensive process, because it takes place in a pusher furnace, which consumes substantial amounts of electricity. Overall, for 1 kg of NMC produced, processes starting from LIB disassembly up to solvent extraction consume 0.12kWh electricity, co-precipitation consumes 0.07kWh electricity, and calcination consumes 7.6kWh electricity. All processes collectively consume 0.93m^3 natural gas/kg NMC produced (Xie *et al* 2015).

The LCIs for NMC precursors production are summarized in Table 3, and the LCIs for NMC production are summarized in Table 4. Note that the LCIs for precursors production are normalized to per ton of precursor produced. Also note that process water consumption is all attributed to precursor production, since the calcination process does not use water. In addition, natural gas use is also attributed to precursor production, since the calcination process is powered by electricity.

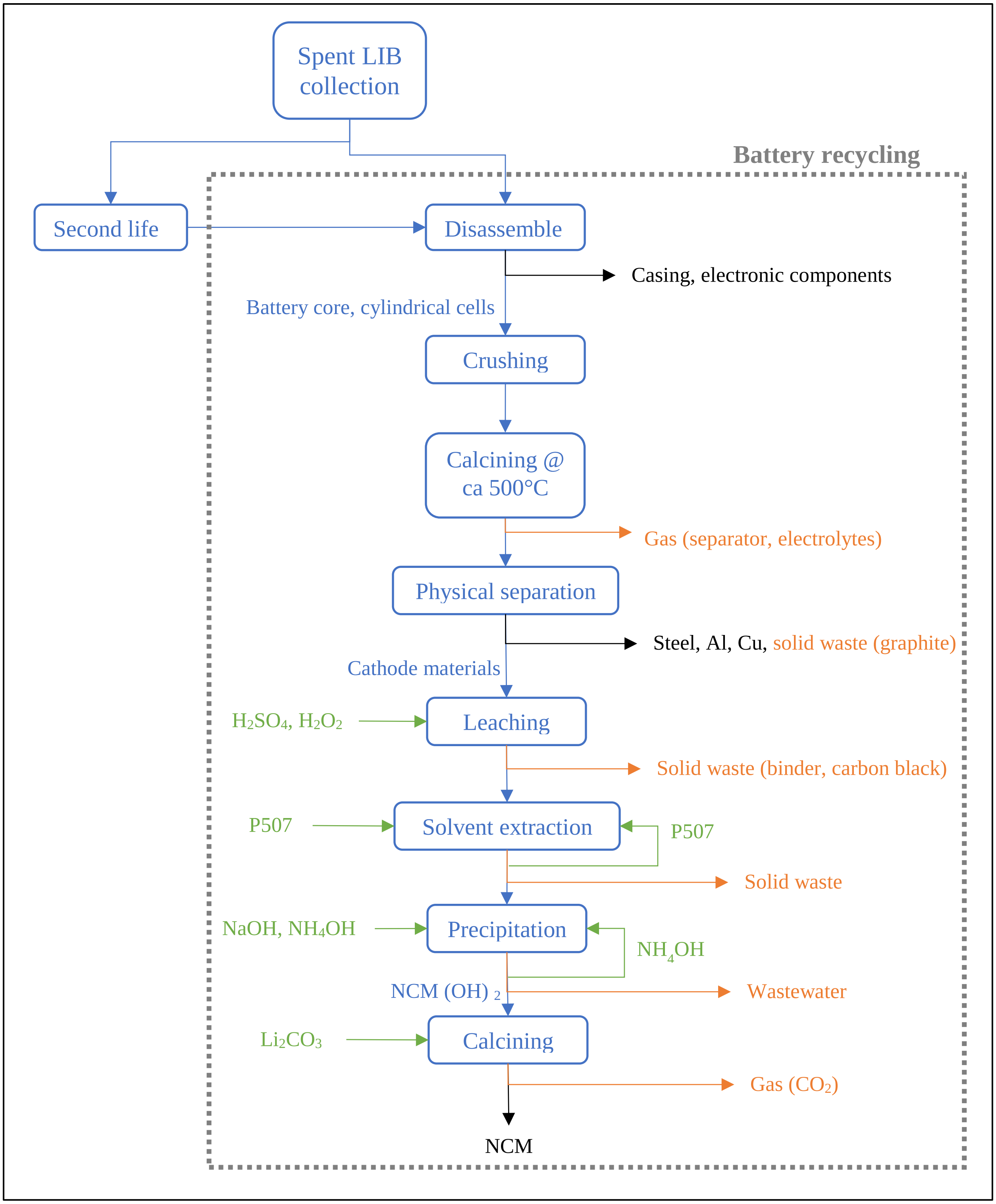


Figure 2 Process flow diagram for NCM production from hydrometallurgical recycling of spent LIB

Table 3. LCIs for NMC precursors production

	GREET2016	GREET2017				
	NMC(424)	NMC(424)	NMC(111)	NMC(622)	NMC(811)	Recycled NMC*
Material inputs (ton/ton NMC precursor						
NiSO4	0.678	0.798	0.663	1.187	1.577	---
MnSO4	0.34	0.4	0.664	0.396	0.197	---
CoSO4	0.662	0.779	0.647	0.386	0.192	---
NaOH	0.877	0.877	0.874	0.869	0.866	1.971
NH_4OH	0.081	0.081	0.081	0.081	0.081	0.110
H_2SO_4	---	---	---	---	---	3.787
HCl	---	---	---	---	---	0.042
H_2O_2	---	---	---	---	---	1.286
Na_2CO_3	---	---	---	---	---	0.074
Process water use (gallon/ton NMC precursor)						
	---	3,853	3,853	3,853	3,853	3,367
Energy inputs (mmbtu/ton NMC precursor)						
Electricity	0.137	0.497	0.497	0.497	0.497	0.621
Natural gas	8.637	2.638	2.638	2.638	2.638	30.974
Total	8.774	3.135	3.135	3.135	3.135	31.595

*Kerosene and P507 use for solvent extraction is minimal and therefore not included.

Table 4. LCIs for NMC production

	GREET2016	GREET2017				
	NMC(424)	NMC(424)	NMC(111)	NMC(622)	NMC(811)	Recycled NMC
Material inputs (ton/ton NMC)						
Precursor	0.949	0.949	0.949	0.949	0.949	0.949
LiOH	0.249	---	---	---	---	---
Li_2CO_3	---	0.4	0.4	0.4	0.4	0.403
O_2	0.083	0.083	0.083	0.083	0.083	---
Energy inputs (mmbtu/ton NMC)						
Electricity	1.88	9.144	9.144	9.144	9.144	23.454
Total	1.88	9.144	9.144	9.144	9.144	23.454
Non-combustion process emissions (g/ton NMC precursor)						
CO2	N/A	294,227*	294,227*	294,227*	294,227*	202,608

*Estimated based on stoichiometry of Li_2CO_3 thermal decomposition.

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