

Life cycle assessment of hydrometallurgical lithium-ion battery recycling processes – evaluating geographical location sensitivity

R.F. Maritz, G. Akdogan and C. Dorfling

Stellenbosch University, South Africa

End-of-life lithium-ion batteries are one of the fastest-growing solid waste streams globally. Hydrometallurgical processes are commonly considered for metal recycling from the cathode material recovered from these batteries. Comparing the environmental impact of processing options plays an important role in process selection and design optimisation. There is an increased need for regional-specific impact assessments considering the influence of local conditions on the environmental performance of processes. The aim of this paper was to compare the environmental impact of hydrometallurgical lithium-ion battery recycling processes in a South African context with the impact of these processes in a European context. The study considered differences in the production pathways of chemical reagents used in the leaching, solution purification and metal recovery steps of the hydrometallurgical processes as well as differences in energy generation technologies between South Africa and Europe. The results showed that the largest difference between the environmental impact of hydrometallurgical recycling of end-of-life lithium-ion batteries in South Africa and Europe is caused by differences in energy generation technologies. This difference in energy generation results in hydrometallurgical processes causing between 77% and 1214% more emissions in South Africa than in the average European country when recycling an equivalent quantity of lithium-ion batteries. Subsequently, a multi-factor environmental analysis was performed to determine that hydro-, wind-, solar thermal-, photovoltaic-, and nuclear power are the most environmentally friendly energy generation methods alongside the combustion of biogas and biomass to replace South Africa's consumption of hard coal.

Keywords: Lithium-ion Batteries, Recycling, Hydrometallurgy, Life Cycle Assessment, Europe, South Africa

INTRODUCTION

Since the turn of the 21st century lithium-ion batteries (LIBs) have become commonplace in both first- and third world countries for energy storage applications in consumer electronics such as laptops, cell phones, and a variety of handheld devices. Alongside the increased use of LIBs, the need for their recycling has also become an important issue. According to a study by Gaines *et al.*, (2023), the global recycling rate for LIBs in 2019 was equal to approximately 59%, with the European Union planning to increase its collection of portable battery waste to 73% by 2030 (Council of the European Union, 2023). In contrast, developing countries such as South Africa are behind global trends in terms of LIB recycling as evidenced by the fact that South Africa had only a recycling rate of roughly 1% in 2019 (Gericke *et al.*, 2019). South Africa is, however, set to experience a large increase in LIB consumption because of the Renewable Energy Independent Power Procurement Programme having started large-scale battery procurement in 2022 (Department of Science and Innovation South Africa, 2023).

This is evidenced by South Africa spending \$1.1 billion on importing lithium-ion batteries in the first six months of 2023 to complement the import of \$650 million worth of solar panels during the same period (Kuhudzai, 2023; Maseko *et al.*, 2023).

As a result, South Africa will have to start rapidly expanding its battery collection and recycling infrastructure to prevent a build-up of e-waste materials in landfill sites. Regarding the recycling of end-of-life LIBs, emerging markets such as South Africa are likely to primarily make use of hydrometallurgy to recover the metals of interest from the LIB waste through leaching and a variety of aqueous recovery strategies. This is because hydrometallurgical recycling processes are more suited to smaller-scale operations (required because of lower recycling rates) and can more easily be expanded in capacity than the competing pyrometallurgical processes, (Chagnes and Swiatowska, 2015). It is, however, unclear whether South Africa will be able to sustainably implement these hydrometallurgical end-of-life LIB recycling processes. It is therefore necessary to implement an established environmental analysis method, such as life cycle assessment (LCA), to study LIB recycling systems (Tolomeo *et al.*, 2020) by quantifying and comparing trade-offs between multiple environmental impacts (Lee and Inaba, 2004). As such, this study focuses on using LCA to calculate and compare the environmental impact of different hydrometallurgical processes used for end-of-life LIB recycling in both developed (Europe) and developing (South Africa) regions.

Methodology

Goal and scope

The goal of this attributional LCA study is to compare the average environmental performance of hydrometallurgical end-of-life LIB recycling processes across the entirety of Europe and South Africa. The study considers a variety of process options, including the use of three different lixiviants and two different metal recovery strategies. This allows the LCA results to be evaluated across several possible scenarios to help validate the conclusions made from this investigation. Furthermore, the system under consideration is a recycling system with one waste stream (LIB waste) entering and several co-products leaving in the form of reclaimed metals. In accordance with a similar study done by Rajaeifar *et al.*, (2021) regarding pyrometallurgical recycling of end-of-life LIBs, the LIB waste entering the LCA system is chosen to be the reference flow. As such, the functional unit is defined as 1 kWh of mixed end-of-life LIB batteries being fed to the LIB recycling process, where the end-of-life LIB mixture has the composition of the global LIB market. The functional unit is defined as an energy flow in agreement with previous LCA literature regarding LIB recycling (Tolomeo *et al.*, 2020) and the co-products formed during recycling are allocated according to the avoided burden approach which treats co-products formed in the LCA system as spared emissions and involves subtracting the environmental impacts of the primary production process from the LCA recycling system under investigation (Nakatani, 2014). During the process simulation, this investigation considers a snapshot of the steady state operation of an end-of-life hydrometallurgical LIB recycling process modelled according to literature values and not primary data that was collected from real operational processes.

Scenarios and system boundaries

The boundaries for the LCA system being modelled during this investigation can be seen in Figure 1, where the pretreatment section is omitted from the LCA system boundary since it has an equal environmental impact for all six of the process options being modelled. Subsequently, the underflow of the flotation process (consisting of mostly cathode material and smaller amounts of graphite-anode and other metallic values) is fed to leaching tanks. For this study, the three lixiviant systems modelled include the hydrochloric, sulphuric, and citric acid systems. Ultimately, the two strategies to recover metals from the pregnant leach solution (PLS) that were modelled during this investigation include the precipitation of a mixed nickel-manganese-cobalt (NMC) product and a hybrid sequential precipitation-solvent extraction (SX) process. The mixed NMC precipitation involves adjusting the ratio of nickel, manganese, and cobalt in the PLS to then precipitate a mixed NMC hydroxide product to be used when resynthesising new NMC cathodes. Meanwhile, the sequential recovery processes (indicated as the SX processes) involve recovering the MOIs in the PLS using specialised chemicals. For the mineral acid processes, dissolved manganese is recovered using potassium permanganate, nickel and cobalt are separated using solvent extraction, the nickel is subsequently precipitated using sodium hydroxide, and the cobalt is either precipitated as an oxalate or the cobalt is precipitated as a sulphate salt. Meanwhile,

for the citric acid process, the nickel and cobalt are separated using dimethylglyoxime, cobalt is precipitated as an oxalate, nickel is precipitated using sodium hydroxide, and manganese is recovered using solvent extraction. Finally, both the mixed NMC and SX processes conclude by subjecting the PLS to forced circulation evaporation upon which the remaining PLS is concentrated, and bulk amounts of sodium salts are precipitated prior to lithium precipitation. For the mineral acid system, the lithium is precipitated from solution using sodium carbonate, whereas the citric acid process utilises phosphate precipitation. Both the mass- and energy balances as well as the full literature review for the process modelling can be provided upon further request (Maritz, 2024).

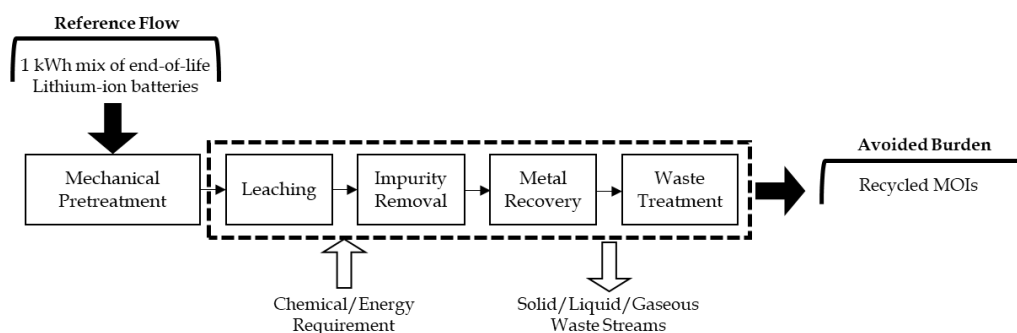


Figure 1. Flow diagram demarcating the scope of the life cycle assessment study.

Life cycle inventory

Process feed considerations

For the purposes of this LCA study, the feed composition of the LIB waste being recycled was defined to be representative of the global LIB market. As such, the global LIB market was approximated to 43% $\text{LiNi}_{0.33}\text{Mn}_{0.33}\text{Co}_{0.33}\text{O}_2$ (NMC₁₁₁), 21% LiCoO_2 (LCO), and 36% LiFePO_4 (LFP) (Gupta and Paranjape, 2021). These fractions were then used to convert the mass of LIB waste being recycled to energy flows (as discussed in the *Goal and scope*) using the energy densities of the different lithium-ion batteries. Accordingly, the energy densities used for NMC₁₁₁, LCO, and LFP are 130 Wh/kg, 203 Wh/kg, and 108 Wh/kg, respectively (Chagnes and Swiatowska, 2015).

Process modelling

The life cycle inventory (LCI) datasets were compiled for each of the six recycling processes detailed in the scenarios and system boundaries section. The foreground data for these inventories were compiled through extensive literature review and generated through mass and energy balances in HSC Chemistry (version 9.07). During this study, the assumption was made that power-to-heat technologies (with the existing electricity grid) would need to be utilised to provide the necessary process heat requirements. The complete inventories for the mixed NMC precipitation and the integrated sequential precipitation - SX processes are provided in Table I and Table II, respectively. Process information related to the order of operations, chemical precipitants, and literature references for these precipitation/SX reactions can be provided upon further request (Maritz, 2024). Furthermore, the background data (regarding electricity mixes, raw material production, and waste treatment) were obtained from the LCA for Experts (previously GaBi) Professional database. The background data was either gathered from existing datasets or generated by combining different existing datasets in stoichiometric ratios (Maritz, 2024).

Life cycle impact assessment and interpretation

The impact assessment section of this LCA investigation makes use of the 2016 ReCiPe (H) method excluding biogenic carbon. The ReCiPe method is a global midpoint and endpoint impact evaluation method that was adapted from both the CML-IA and outdated Eco-Indicator 99 method in 2000 to create a system which completely harmonises the use of endpoint and midpoint indicators. The life cycle impact assessment for this study was executed using software from Sphera called LCA for Experts (previously GaBi – content version 10.7.1.28) which has been used with notable success in previous LCA studies regarding LIB recycling (Tolomeo *et al.*, 2020). The four impact categories selected for this impact assessment are climate change, freshwater consumption, land use, and terrestrial acidification. Both

acidification and climate change were chosen based on their popularity in previous LCA studies regarding LIB manufacturing and recycling (Tolomeo *et al.*, 2020) while the remaining impact categories were selected to represent the impact categories that have traditionally been important in a South African environmental context (Brent, 2004). The interpretation of the impact assessment data consists of comparative analysis between the European and South African contexts by using the formula shown in Equation 1, where Y indicates the impact category under consideration and ΔY is the percentage change in the specific impact category when changing from the European to the South African scenario. If the value for ΔY is negative, then the South African scenario is more environmentally friendly than the European scenario and if the value for ΔY is positive, then the inverse holds true.

$$\Delta Y = (Y_{ZA} - Y_{EU}/|Y_{EU}|) \times 100 \quad [1]$$

Table I. LCI data generated for the treatment of 1 kWh LIB waste using mixed NMC precipitation processes

HCl	Total	Unit	H ₂ SO ₄	Total	Unit	C ₆ H ₈ O ₇	Total	Unit
Process Feed	IN		Process Feed	IN		Process Feed	IN	
LIB waste	1	kWh	LIB waste	1	kWh	LIB waste	1	kWh
LCO	1.51	kg	LCO	1.51	kg	LCO	1.51	kg
LFP	2.64	kg	LFP	2.64	kg	LFP	2.64	kg
NMC ₁₁₁	3.14	kg	NMC ₁₁₁	3.14	kg	NMC ₁₁₁	3.14	kg
Reactants			Reactants			Reactants		
HCl	55.4	kg	H ₂ SO ₄	37.3	kg	C ₆ H ₈ O ₇	72.9	kg
NaOH	60.5	kg	H ₂ O ₂	21.9	kg	H ₂ O ₂	22	kg
MnSO ₄	2.1	kg	NaOH	30.4	kg	NaOH	44.9	kg
NiSO ₄	2.3	kg	MnSO ₄	2.2	kg	MnSO ₄	2.3	kg
Na ₂ CO ₃	3.1	kg	NiSO ₄	2.3	kg	NiSO ₄	2.3	kg
			Na ₂ CO ₃	3	kg	NaH ₂ PO ₄	8.6	kg
Energy			Energy			Energy		
Electricity	190.2	kWh	Electricity	131	kWh	Electricity	185.3	kWh
Water			Water			Water		
Process	184.7	kg	Process	270.9	kg	Process	391.5	kg
Distilled	295	kg	Distilled	146.3	kg	Distilled	133.5	kg
Waste	OUT		Waste	OUT		Waste	OUT	
Landfill	1.6	kg	Landfill	1.5	kg	Landfill	0.4	kg
Wastewater	4.7	kg	Wastewater	31.5	kg	Wastewater	58.5	kg
Avoided products		kg	Avoided products		kg	Avoided products		kg
Ni(OH) ₂	2.3	kg	Ni(OH) ₂	2.3	kg	Ni ₃ (PO ₄) ₂	3.1	kg
Mn(OH) ₂	2.2	kg	Mn(OH) ₂	2.2	kg	Mn ₃ (PO ₄) ₂	3	kg
Co(OH) ₂	2.3	kg	Co(OH) ₂	2.3	kg	Co ₃ (PO ₄) ₂	3.1	kg
Li ₂ CO ₃	2.2	kg	Li ₂ CO ₃	2.1	kg	Li ₃ PO ₄	2.4	kg
FePO ₄	2.2	kg	FePO ₄	2.5	kg	FePO ₄	2.4	kg
NaCl	80.6	kg	Na ₂ SO ₄	48.9	kg	Na ₃ C ₆ H ₅ O ₇	73.2	kg

Table II. LCI data generated for the treatment of 1 kWh LIB waste using integrated precipitation/SX processes

HCl	Total	Unit	H ₂ SO ₄	Total	Unit	C ₆ H ₈ O ₇	Total	Unit
Process Feed	<u>IN</u>		Process Feed	<u>IN</u>		Process Feed	<u>IN</u>	
LIB waste	1.0	kWh	LIB waste	1.0	kWh	LIB waste	1.0	kWh
LCO	1.51	kg	LCO	1.51	kg	LCO	1.51	kg
LFP	2.64	kg	LFP	2.64	kg	LFP	2.64	kg
NMC ₁₁₁	3.14	kg	NMC ₁₁₁	3.14	kg	NMC ₁₁₁	3.14	kg
Reactants			Reactants			Reactants		
HCl	55.7	kg	H ₂ SO ₄	61.7	kg	C ₆ H ₈ O ₇	72.9	kg
H ₂ SO ₄	2.8	kg	H ₂ O ₂	21.9	kg	H ₂ O ₂	22.0	kg
NaOH	56.0	kg	NaOH	25.2	kg	NaOH	50.5	kg
KMnO ₄	3.4	kg	KMnO ₄	2.0	kg	C ₄ H ₈ N ₂ O ₂	0.2	kg
PC-88A	1.2	kg	Cyanex 272	0.8	kg	HCl	0.8	kg
C ₁₂ H ₂₆ C ₁₅ H ₃₂	2.3	kg	C ₁₂ H ₂₆ C ₁₅ H ₃₂	2.6	kg	H ₂ SO ₄	6.3	kg
CoCl ₂	1.3	kg	Na ₂ CO ₃	3.0	kg	(NH ₄) ₂ C ₂ O ₄	3.9	kg
(NH ₄) ₂ C ₂ O ₄	4.0	kg	Energy			H ₃ PO ₄	2.0	kg
Na ₂ CO ₃	2.6	kg	Electricity	539.5	kWh	Na ₂ CO ₃	13.4	kg
Energy			Water			Na-D ₂ EHPA	2.5	kg
Electricity	207.9	kWh	Process	615.5	kg	C ₁₂ H ₂₆ C ₁₅ H ₃₂	7.3	kg
Water			Distilled	132.5	kg	C ₁₂ H ₂₇ O ₄ P	0.6	kg
Process	342.3	kg				Energy		
Distilled	286.5	kg				Electricity	437.4	kWh
						Water		
						Process	872.4	kg
						Distilled	144.6	kg
Waste	<u>OUT</u>		Waste	<u>OUT</u>		Waste	<u>OUT</u>	
Landfill	3.0	kg	Landfill	2.0	kg	Landfill	0.4	kg
Wastewater	57.7	kg	Wastewater	80.6	kg	Wastewater	239.5	kg
Avoided products			Avoided products			Avoided products		
Ni(OH) ₂	0.9	kg	Ni(OH) ₂	0.9	kg	Ni(OH) ₂	1.0	kg
Mn(OH) ₂	0.1	kg	Mn(OH) ₂	0.1	kg	Mn(OH) ₂	0.9	kg
MnO ₂	1.5	kg	MnO ₂	1.3	kg	CoC ₂ O ₄	3.7	kg
CoC ₂ O ₄	4.7	kg	CoSO ₄	3.7	kg	Li ₂ CO ₃	2.4	kg
Li ₂ CO ₃	1.8	kg	Li ₂ CO ₃	2.1	kg	FePO ₄	2.4	kg
FePO ₄	1.4	kg	FePO ₄	2.5	kg	Na ₃ C ₆ H ₅ O ₇	57.7	kg
NaCl	76.4	kg	Na ₂ SO ₄	39.2	kg			

Results and discussion

Comparison of geographical regions

The first results produced during this study are the LCA results for the European and South African contexts of hydrometallurgical LIB recycling as shown in Table III. It should be noted that negative values indicate that the recycling process has a nett beneficial impact on the environment whereas positive values indicate a nett detrimental impact from recycling. Regarding trends, the mixed NMC

precipitation processes generally outperformed the other metal recovery option while the sulphuric acid processes outperformed other lixiviant systems. From Table III it can also be inferred that hydrometallurgical LIB recycling is much more environmentally friendly in Europe than in South Africa (as reflected in the endpoint values). This is deemed to be the result of larger environmental impacts on acidification, climate change, and freshwater consumption. Despite this trend, the South African processes do have a smaller environmental impact on land use than their European counterparts. The rest of this investigation is focused towards determining the origin of these differences.

Table I. European and South African LCA results for the six LIB recycling processes options considered

Recycling Processes	Acidification (kg SO ₂ eq.)		Climate Change (kg CO ₂ eq.)		Water Use (m ³)		Land Use (Crop eq.*y)		Endpoint Values (H/H)	
	EU	ZA	EU	ZA	EU	ZA	EU	ZA	EU	ZA
HCl NMC	-1.5	0.4	29.5	211.0	-0.7	0.3	5.1	2.2	-68.9	20.0
HCl SX	-1.4	0.7	60.4	258.0	-0.2	0.8	7.0	3.7	-43.7	52.9
H ₂ SO ₄ NMC	-1.4	-0.2	19.6	126.0	-1.0	-0.5	2.0	-2.2	-70.8	-16.9
H ₂ SO ₄ SX	-1.1	4.1	150.0	587.0	0.1	2.0	15.6	-0.8	-18.4	206.0
Citric NMC	-1.4	0.5	130.0	266.0	2.9	3.5	60.2	54.1	-19.8	53.3
Citric SX	-1.2	3.1	248.0	582.0	4.5	6.0	78.8	65.1	32.8	210.0

Contributing factors towards the difference between geographical regions

In the discussion to follow, the environmental impact of the South African context will be shown as a percentage difference (ΔY) from the European context. Again, a positive percentage indicates that the South African process is less environmentally friendly than its European counterpart, whereas a negative value indicates the opposite. The production routes of various chemicals such as hydrochloric acid, sulphuric acid, and sodium hydroxide in Europe and South Africa are first compared, followed by a comparison of the electricity infrastructure of Europe and South Africa.

Hydrochloric- and sulphuric acid

The composition of both the European and South African markets for hydrochloric acid production are reflected in Table IV, accompanied by the comparative environmental impact of using hydrochloric acid produced in South Africa instead of Europe to recycle end-of-life LIBs as shown in Figure 2. Regarding the market compositions, it can be seen from GaBi databases (Sphera Solutions GmbH, 2023a) that Europe produces most of its hydrochloric acid as the co-product of inorganic chlorination and halogen exchange reactions. Europe also produces roughly 8% of its hydrochloric acid by combusting hydrogen and chlorine gas (both produced from electrolysis) to form hydrogen chloride, which is the only route for producing hydrochloric acid in South Africa. There are three forms of electrolysis available to produce the chemicals necessary for hydrochloric acid production, namely: mercury, diaphragm (asbestos or synthetic), and membrane cell electrolysis. The main difference between these processes is the medium used to separate the anode from the cathode, which allows the half reactions in equilibrium to take place, while also allowing sodium ions to permeate through (Crook and Mousavi, 2016). From 2018 data (Valette, 2018), it was calculated that the South African market for hydrochloric acid production consisted of roughly 56.8% Teflon membrane cell electrolysis and 43.2% synthetic diaphragm electrolysis.

Table IV. Market analysis of hydrochloric acid production in Europe and South Africa

EU production		ZA production	
Chlorination and halogen exchange	90.0%	Teflon (membrane) cell electrolysis	56.8%
H ₂ + Cl ₂ (electrolysis)	8.0%	Synthetic (diaphragm) cell electrolysis	43.2%
NaCl+H ₂ SO ₄	2.0%		

To simplify this investigation, it was assumed that the entire South African context can be approximated as hydrochloric acid production from chlorine. The two datasets used to model the background data for each scenario can be seen in the bullet points below and it should be mentioned that the hydrochloric acid flows were adjusted according to the concentration stated by the dataset:

- EU - DE: Hydrochloric acid (100%) Sphera technology mix
- ZA - DE: Hydrochloric acid (32%) Sphera primarily from chlorine

After having modelled the two separate scenarios, the HCl LCA results in Figure 2 were produced. The South African scenario was found to have a significantly more detrimental impact on climate change, water consumption and land use. However, this only causes the South African hydrochloric acid end-of-life LIB recycling processes to perform roughly 2.3% to 3.7% worse overall than their European counterparts (as suggested by the final endpoint values). This simultaneously reflects the relative importance of the midpoint impact categories not considered during this study and the importance of developing a new set of weighting and normalisation factors specific to South Africa. If not, then the importance of water- and land use will be underestimated when using endpoint values to calculate the environmental impact of end-of-life hydrometallurgical LIB recycling in South Africa.

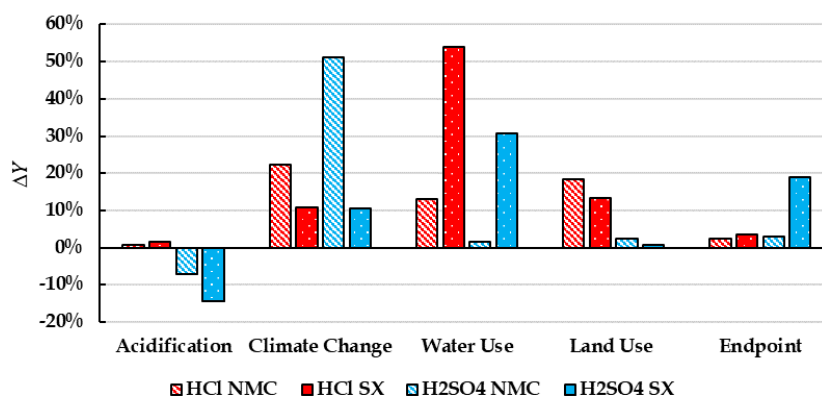


Figure 2. Comparative LCA of LIB recycling using HCl and H₂SO₄ produced in South Africa vs Europe.

Regarding sulfuric acid production, the compositions of both the European and South African markets are reflected in Table V, accompanied by the comparative environmental impact of using sulfuric acid produced in South Africa instead of Europe to recycle end-of-life LIBs also shown in Figure 2. Regarding the market compositions, it can be seen from GaBi databases (Fertilizers Europe, 2024) that Europe produces almost 85% of its sulfuric acid by extracting sulfur impurities from either natural gas, crude oil, or smelter offgas. South Africa however, produces practically all its sulfuric acid from sulfide impurities present in either crude oil or coal (gasified and used as feedstock to the Fischer Tropsch process). Using data from 2012 (Modiselle, 2013), the market composition for sulfuric acid production in South Africa is roughly approximated to originate 53.8% from crude oil production and 46.2% from crude oil desulfurisation.

Table V. Market analysis of sulfuric acid production in Europe and South Africa

EU production		ZA production	
Pyrite burning route	4.7%	Fischer Tropsch (Sasol)	53.8%
Smelter gas (non-ferrous)	37.6%	Crude Oil (Natref, Sapref, Engen Caltex)	46.2%
Natgas/Crude Oil (Sulphur route)	46.1%		
H ₂ SO ₄ regeneration and recovery	11.6%		

The assumption was made for this investigation that all the sulfuric acid used to recycle end-of-life lithium-ion batteries in South Africa would come from Sasol, which is the single biggest supplier of granular sulfur in South Africa. GaBi does, however, not have a dataset for sulfuric acid production from synthetic fuels and thus the sulfuric acid flows were modelled using the datasets shown below:

- EU – RER: Sulfuric acid (100% H₂SO₄) Fertilizers Europe, oxidation of sulfur
- ZA – RER: Sulfuric acid aq. from natural gas (96%) Sphera, oxidation of sulfur

The LCA results in Figure 2 indicate that the South African sulfuric acid production method (via natural gas/synthetic fuel desulfurisation) is 3% to 19% worse overall for recycling end-of-life LIBs when comparing endpoint values. This comes as the result of the South African production method having a significantly more detrimental impact on climate change and water consumption, despite having a significantly less detrimental impact on acidification.

Sodium hydroxide

The composition of both the European and South African markets for sodium hydroxide production are reflected in Table VI, accompanied by the comparative environmental impact of using sodium hydroxide produced in South Africa instead of Europe to recycle end-of-life LIBs as shown in Figure 3. The main process used industrially to produce sodium hydroxide is the chloralkali electrolysis process (Sphera Solutions GmbH, 2023b) which has been discussed in detail during the hydrochloric acid section, which is also why the South African market composition is the same for hydrochloric acid and sodium hydroxide.

Table VI. Market analysis of sodium hydroxide production in Europe and South Africa

EU production		ZA production	
Teflon (membrane) cell electrolysis	60.0%	Teflon (membrane) cell electrolysis	56.8%
Asbestos (diaphragm) cell electrolysis	13.0%	Synthetic (diaphragm) cell electrolysis	43.2%
Mercury cell electrolysis	27.0%		

From previous LCA work (Garcia-Herrero *et al.*, 2017), it can be inferred that membrane cell electrolysis was found to have 92% less environmental burden than the mercury cell electrolysis process and a little more than 25% reduction in environmental burden when compared to the asbestos diaphragm cell electrolysis process (Garcia-Herrero *et al.*, 2017). Using this information and approximating the South African market as being fully operated by membrane cell electrolysis (due to a lack of LCA information regarding synthetic diaphragm cell electrolysis), a conversion factor between the South African and European market can be calculated through normalisation, as shown in Equations 2 and 3, where x is the European market fraction for given electrolysis technologies:

$$\text{Conversion Factor} = x_{\text{membrane}} + 0.08x_{\text{mercury}} + 0.75x_{\text{asbestos}} \cong 0.72 \quad [2]$$

$$\text{NaOH consumption (ZA)} = \text{NaOH consumption (EU)} \times \text{Conversion factor} \quad [3]$$

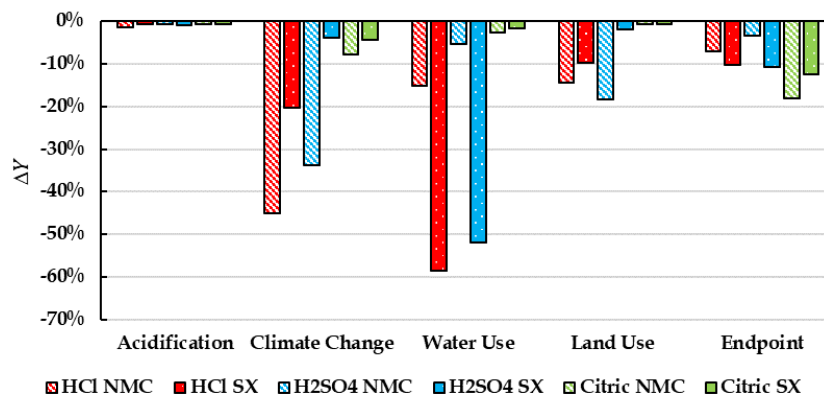


Figure 3. Comparative LCA of LIB recycling using NaOH produced in South Africa vs Europe.

The dataset used for sodium hydroxide production was RER: Sodium hydroxide (caustic soda) mix (100%) Sphera technology mix. Having converted the European context to the South African context shown in Figure 3, it can be seen that the South African sodium hydroxide mix may reduce the environmental impact of end-of-life LIB recycling processes by between 3.4% and 18.2%, depending on process-specific sensitivity to sodium hydroxide consumption.

Electricity

The composition of both the European and South African markets for electricity production are reflected in Table VII, accompanied by the comparative environmental impact of using electricity produced in South Africa instead of Europe to recycle end-of-life LIBs as shown in Figure 4. Regarding the market compositions, the GaBi databases currently indicate that Europe produces more than 50% of its electricity through more sustainable technologies such as hydro power, wind power, natural gas, and nuclear power compared to South Africa's electricity mix which reportedly consists of approximately 90% hard coal consumption according to the GaBi Professional Core database.

Table VII. Market analysis of electricity production in Europe and South Africa generated from GaBi

Sources	Nuclear	Hydro	Natural gas	Wind	Hard coal	Photovoltaic	Fuel oil	Biomass	Lignite
EU	25.3%	11.6%	19.1%	11.6%	9.7%	3.8%	1.7%	3.1%	9.0%
ZA	4.5%	2.2%	-	2.5%	88.8%	1.3%	0.1%	0.2%	-

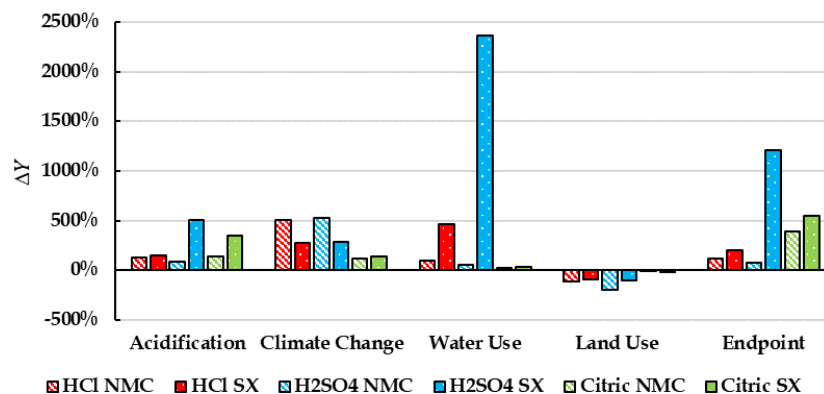


Figure 4. Comparative LCA of LIB recycling using electricity produced in South Africa vs Europe.

Upon considering the comparative environmental performance of the hydrometallurgical end-of-life LIB recycling processes in Figure 4, the South African context is worst in every impact category except land use. Altogether, the South African electricity mix makes the end-of-life LIB recycling process 77% to 1214% less environmentally friendly compared to the European electricity mix. It is thus imperative that South Africa expands its sustainable energy infrastructure.

Heat generation study

This section investigates the different forms of energy generation and determines which are the most environmentally friendly options available for a developing country like South Africa to reduce their reliance on fossil fuels. To this end, a life cycle impact assessment sensitivity investigation was conducted, and the results are shown in Table VIII. The reason for conducting an impact assessment sensitivity investigation is due to the presence of biases in each impact assessment method, and by performing an investigation where several impact assessment methods are considered and assigned equal importance, the effect of bias on the results is minimised.

Table VIII. Summarised endpoint rankings for producing 1 MJ of heat using different energy sources

	Electrical resistive heating (power-to-heat)									On-site thermal combustion heating								
	Coal Gases	Geo-Thermal	Hard Coal	Hydropower	Nuclear power	Photovoltaics	Solar Thermal	Waste to Energy	Wind power	Biogas	Biomass	Hard Coal	Heavy Fuel Oil	Light Fuel Oil	Lignite	LPG	Natural gas	Peat
A	11	14	17	1	2	9	4	16	3	6	18	12	10	7	13	8	5	15
B	17	18	16	1	14	4	3	15	2	7	5	10	12	9	11	8	6	13
C	18	9	17	1	3	7	4	8	6	5	2	11	13	15	12	14	10	16
D	18	14	17	1	15	4	3	16	2	7	5	8	13	11	9	10	6	12
E	16	18	17	3	2	6	5	15	4	1	7	8	14	13	9	12	11	10
F	17	18	16	3	1	2	7	15	6	13	4	8	12	11	10	9	5	14
∑	97	91	100	10	37	32	26	85	23	39	41	57	74	66	64	61	43	80
#	17	16	18	1	5	4	3	15	2	6	7	9	13	12	11	10	8	14

A=CML B=EF3 C=EPS D=ReCiPe E=TRACI F=UBP

This investigation includes several European methods such as CML-IA, Environmental Footprint, Environmental Priority Strategies, and the Ecology Scarcity (UBP) method, followed by an American perspective using the TRACI method and finally a global perspective using the ReCiPe method (Acero *et al.*, 2016). Take note of the abbreviation for liquified petroleum gas (LPG). Upon investigation of Table VIII, the top seven most environmentally friendly methods of energy generation are hydro power, wind power, solar thermal power, photovoltaic power, nuclear power, and energy produced from the on-site combustion of biogas and biomass. South Africa has primarily made use of on-site combustion of natural gas to fulfil its needs for process heat in recent years but is facing a looming supply shortage (Mahlaka, 2024). If South Africa can sustainably source alternative fuels (e.g., biogas and biomass) to provide heat through on-site combustion, then this would also be preferable to using on-site combustion of natural gas and to the implementation of power-to-heat technology using South Africa's current energy infrastructure, considering its dependence on hard coal.

Data quality

It is important to note that several data resources used during this study are five to ten years old and that little information is freely available to the public to be able to assess the current nature of chemical production in South Africa. Furthermore, the data that is available often needs to be bought and even then, may be questionable as shown by the fact that the GaBi database reports South Africa as producing 90% of its energy from hard coal whereas the latest data from the International Energy Agency reports

South Africa producing only 70% of its energy from hard coal (International Energy Agency, 2024). The onus is thus on both the South African public- and private sector to ensure that this type of information is accurate and freely available to the public. Without sufficient market data South Africa will have no way to prove to the global economy that they are viable contenders in LIB recycling and other circular economic activities. It is also recommended that a uniquely South African environmental impact assessment method may be developed to showcase the unique environmental challenges faced by the South African chemical process industry to avoid unfair comparison to Europe and the United States when performing LCA and other environmental impact studies.

ACKNOWLEDGEMENTS

The work presented was supported financially by the Department of Science and Innovation (DSI), associated with the ERA-MIN2 programme (ELiMINATE project). Further acknowledgement is made towards the Wilhelm Frank Trust for their financial support.

CONCLUSIONS AND RECOMMENDATIONS

In conclusion, this investigation aimed to compare the environmental impact of hydrometallurgical LIB recycling facilities in Europe and facilities in South Africa to assess the sustainability of future LIB recycling facilities in South Africa. The study compared the industrial production pathways of hydrochloric acid, sulphuric acid, and sodium hydroxide produced in the different geographical regions. The study also considered differences in the energy generation infrastructure in Europe and South Africa. The investigation showed that main distinguishing factor between hydrometallurgical LIB recycling in Europe and South Africa is the difference in energy production, causing 77% to 1214% more emissions in South Africa than in Europe for LIB recycling. This is because Europe produces more than 50% of their energy through sustainable process technologies, whereas South Africa is reliant on the combustion of hard coal to produce most of its process energy. Lastly, the importance of accurate South African market data is emphasised if South Africa is to form part of the global circular economy.

REFERENCES

- Acero, A.P., Rodríguez, C., Changelog, A.C., 2016. LCIA methods Impact assessment methods in Life Cycle Assessment and their impact categories.
- Brent, A.C., 2004. Development of a Life Cycle Impact Assessment procedure for Life Cycle Management in South Africa (PhD). University of Pretoria, Pretoria.
- Chagnes, A., Swiatowska, J., 2015. Lithium process chemistry: Resources, extraction, batteries, and recycling, 1st ed, Lithium Process Chemistry: Resources, Extraction, Batteries, and Recycling. Elsevier, Amsterdam, Netherlands. <https://doi.org/10.1016/C2013-0-19081-2>
- Council of the European Union, 2023. Council adopts new regulation on batteries and waste batteries. Brussels.
- Crook, J., Mousavi, A., 2016. The chlor-alkali process: A review of history and pollution. Environ Forensics. <https://doi.org/10.1080/15275922.2016.1177755>
- Department of Science and Innovation South Africa, 2023. Draft South African Renewable Energy Masterplan (SAREM). Pretoria.
- Fertilizers Europe, 2024. Process data set: Sulphuric acid (100% H₂SO₄); oxidation of sulphur, including primary production; production mix, at plant; 100 % (en) [WWW Document]. URL <https://sphera.com/2023/xml-data/processes/0fb7d4a6-2d72-4403-84b8-83b6788ef2db.xml> (accessed 1.24.24).

- Gaines, L., Zhang, J., He, X., Bouchard, J., Melin, H.E., 2023. Tracking Flows of End-of-Life Battery Materials and Manufacturing Scrap. *Batteries* 9. <https://doi.org/10.3390/batteries9070360>
- Garcia-Herrero, I., Margallo, M., Onandía, R., Aldaco, R., Irabien, A., 2017. Life Cycle Assessment model for the chlor-alkali process: A comprehensive review of resources and available technologies. *Sustain Prod Consum* 12, 44–58. <https://doi.org/10.1016/j.spc.2017.05.001>
- Gericke, M., And, W., Robertson S, 2019. Technology landscape report and business case for the recycling of Li-ion batteries in South Africa. Pretoria.
- Gupta, A., Paranjape, N., 2021. Lithium Ion Battery Market Outlook 2020-2026 | Share Analysis [WWW Document]. Lithium Ion Battery Market By Chemistry (LFP, LCO, LTO, NMC, NCA, LMO), By Component (Cathode, Anode, Separators, Electrolytes, Aluminum Foil, Copper Foil, Others), By Application (Industrial {Military, Industrial Equipment, Medical, Marine, Telecommunica. URL <https://www.gminsights.com/industry-analysis/lithium-ion-battery-market> (accessed 4.22.21).
- International Energy Agency, 2024. South Africa - Countries & Regions - IEA [WWW Document]. URL <https://www.iea.org/countries/south-africa> (accessed 8.13.24).
- Kuhudzai, R.J., 2023. South Africa Imported \$1.1 Billion (4.4 GWh) Of Lithium-Ion Cells & Batteries In First 6 Months Of 2023! [WWW Document]. Clean Technica. URL <https://cleantechnica.com/2023/08/05/south-africa-imported-1-1-billion-4-4-gwh-of-lithium-ion-cells-batteries-in-first-6-months-of-2023/> (accessed 5.6.24).
- Lee, K.-M., Inaba, A., 2004. Life Cycle Assessment: Best Practices of International Organization for Standardization (ISO) 14040 Series, Committee on Trade and Investment. Sejong-si, Republic of Korea.
- Mahlaka, R., 2024. SA has ‘four months’ to avoid a natural gas Day Zero as Sasol contract supply crisis looms [WWW Document]. Daily Maverick. URL <https://www.dailymaverick.co.za/article/2024-02-27-sa-has-four-months-to-avoid-a-natural-gas-day-zero-as-sasol-contract-supply-crisis-looms/> (accessed 5.23.24).
- Maritz, R.F., 2024. Comparing the environmental impact of different hydrometallurgical processes for the recycling of lithium-ion batteries using a life cycle assessment approach (Doctoral). University of Stellenbosch, Stellenbosch.
- Maseko, N., Mthembu, L., Mashiane, K., Ramos, D., Levin, S., 2023. TIPS Import tracker second quarter 2023. Pretoria.
- Modiselle, M., 2013. Review of the sulphur industry in the Republic of South Africa 2012. Pretoria.
- Nakatani, J., 2014. Life Cycle Inventory Analysis of Recycling: Mathematical and Graphical Frameworks. *Sustainability* 6, 6158–6169. <https://doi.org/10.3390/su6096158>
- Rajaeifar, M.A., Raugei, M., Steubing, B., Hartwell, A., Anderson, P.A., Heidrich, O., 2021. Life cycle assessment of lithium-ion battery recycling using pyrometallurgical technologies. *J Ind Ecol* 25, 1560–1571. <https://doi.org/10.1111/jiec.13157>
- Sphera Solutions GmbH, 2023a. Process data set: Hydrochloric acid mix (100%); technology mix; consumption mix, to consumer; 100% HCl (en) [WWW Document]. Sphera Managed LCA Content Databases (MLC). URL <https://lcadatabase.sphera.com/2024/xml-data/processes/b80355ff-d10c-42bb-a0af-49dea028c527.xml> (accessed 1.24.24).
- Sphera Solutions GmbH, 2023b. Process data set: Sodium hydroxide (caustic soda) mix (100%); technology mix; production mix, at plant; 1 (en) [WWW Document]. Sphera Managed LCA Content Databases (MLC). URL <https://lcadatabase.sphera.com/2024/xml-data/processes/8034f0ff-d0b9-47a8-86e8-ece772f56871.xml> (accessed 5.7.24).

Tolomeo, R., De Feo, G., Adami, R., Osséo, L.S., 2020. Application of life cycle assessment to lithium ion batteries in the automotive sector. *Sustainability* (Switzerland) 12. <https://doi.org/10.3390/su12114628>

Valette, J., 2018. Chlorine and Building Materials A Global Inventory of Production Technologies, Markets, and Pollution Phase 1: Africa, The Americas, and Europe.



Dr Roelof Frederick Maritz

Postdoctoral Research Fellow
University of Stellenbosch

Dr Maritz is a researcher for the Department of Chemical Engineering at Stellenbosch University. His research focuses on performing the process modelling and the life cycle assessment studies for hydrometallurgical lithium-ion battery recycling processes. He graduated from Stellenbosch with his PhD in March 2024 for the submission of a thesis entitled “Comparing the environmental impact of different hydrometallurgical processes for the recycling of lithium-ion batteries using a life cycle assessment approach”.

