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Comparative Life Cycle Assessment of Lithium-Ion Capacitors Production from Primary Ore and Recycled Minerals

Using LCA to balance environmental, economic and social performance in early phase research and development

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The life cycle assessment (LCA) methodology which allows quantification of environmental performance of products and processes based on complete product life cycle was utilised to evaluate the environmental burdens associated with manufacturing a 48 V lithium-ion capacitor (LIC) module. The prospective LCA compared the environmental impact of manufacturing a LIC module using primary ore materials and recycled materials from end-of-life LICs. For both the primary ore and recycled materials processes, the anode preparation stage was associated with the majority of the climate change and terrestrial acidification burdens. LIC module production utilising recovered materials from end-of-life LICs reduced the environmental impact compared to utilisation of primary ore resources. Application of the LCA methodology in early phase research and development (R&D) activities was demonstrated with a case study on reagent choice decision-

making process that accounted for environmental impact, technical performance and costs in alignment with the sustainability triple bottom line concept.

1. Introduction

Growing environmental concerns have made it imperative to reduce global climate change and this has resulted in prolific development of various energy storage technologies for different applications ranging from portable electronic devices (PED) to electric vehicles (EVs) (1, 2). The most common chemical energy storage devices are batteries for applications requiring high energy density and electrochemical capacitors (ECs) for applications with high power density requirements (3–5). LICs which have the combined desirable properties of batteries (high energy density) and ECs (high power density) are increasingly being investigated as high-performance energy storage devices that have a significant role in the decarbonisation of the transport sector (6, 7).

While there are many promising negative electrode materials for LICs, the lithium titanium oxide ($\text{Li}_4\text{Ti}_5\text{O}_{12}$, LTO) based anode offers high stability towards charge-discharge cycles, faradaic efficiency and lower costs (8–10). As the envisaged use of the LTO based LIC is in hybrid and EVs to assist in decarbonising the transport sector, it becomes pertinent to conduct a LCA for the production of a LIC using primary ore minerals and make comparisons to a manufacturing process that relies on recycling end-of-life LIC. LCA is defined as

a process to evaluate the environmental burdens associated with a product, process or activity by identifying and quantifying energy and materials used and wastes released to the environment (11). The assessment includes the entire life cycle of a product, process or activity, encompassing extracting and processing raw materials, manufacturing, transport and distribution, use, reuse, recycling and final disposal.

LCA facilitates informed decision making as comparative analysis of competing processes or products can be conducted based on environmental impact. At the early stages of R&D activities, LCA is an invaluable tool as it can inform process and material choices that support sustainability goals in addition to promoting innovation for designing products that are more amenable to recycling when they reach end-of-life (12–14). Increasingly, LCA is also being utilised to engage with stakeholders as an evolving green marketing tool through brand competitive differentiation on the basis of sustainability as well as regulatory compliance purposes (15–17). Besides the multifaceted benefits of LCA, its utilisation is not without limitations with uncertainties in inventory data, methodology and application of the weighting technique often being cited as major weaknesses of the approach (18, 19).

While the LCA methodology has been widely applied to energy storage systems this has mostly been for lithium-ion batteries (LIBs), with most studies having focussed on comparative analysis of LIBs to internal combustion engine (ICE) or sustainability of the different battery chemistries (20–22). There is a scarcity in the literature of LCA studies that have analysed production of energy storage devices using primary ore materials in comparison to manufacture of a similar product using recycled materials and specifically for LICs. This study objective is to take a comparative approach with the aim of utilising LCA to inform early phase R&D activities to improve the sustainability of the various process and reagents choices in the production of a LIC module.

The LCA study was conducted as part of the Advanced Lithium Ion Capacitors Electrodes (ALICE) project whose objective was to develop a 48 V LIC module for use in automotive, e-bus and materials handling equipment. The project consortium had industrial and academic partners for developing and scaling-up materials production including application of novel coating techniques to electrode structure to improve performance. The 48 V module built in the project was tested

based on end user requirements and physics based numerical modelling applied at different stages of the project to interlink sophisticated layer structure characterisation results with cell performance.

2. Methodology

2.1 Goal and Scope

The goal of this study is to evaluate the environmental impact of manufacturing a LIC using primary ore materials and making comparative studies for LIC module manufacture using recycled materials from an end-of-life LIC. The scope which captures the pertinent choices for the study is execution of the LCA on the basis of a cradle-to-gate manufacturing process of a 48 V LIC module. The cradle-to-gate approach was considered sufficient given that the goal of the study was for a comparative analysis of LICs production processes from primary ore and recycled minerals. The other stages of LIC product life once the manufactured product is at the gate would be expected to be similar for purposes of making a fair comparison and therefore their exclusion should not affect the results with respect to the goal of this LCA study.

2.2 Functional Unit and System Boundary

The functional unit, which defines the basis for comparison, is the cells that make the 48 V LIC module. The choice of the functional unit was based on capturing the environmental burdens that would make a difference for LICs production processes from primary ore or recycled minerals. The choice is also additionally informed by the potential application of the LIC in hybrid vehicles and therefore cells which make a 48 V LIC module considered an appropriate functional unit. The system boundaries using primary ore materials and production of a LIC module using recycled materials from an end-of-life LIC are shown in **Figure 1** and **Figure 2**. The system boundary includes raw material extraction, electrode material production and cell build for the 48 V LIC module. Both system boundaries exclude the operational usage stage as the attributable environmental burden for this stage would be identical whether a LIC was manufactured using primary (ore) materials or recycled materials from an end of life LIC. As the project consortia members did not have a mechanical disassembler, the system boundary chosen for the recycled materials study and shown

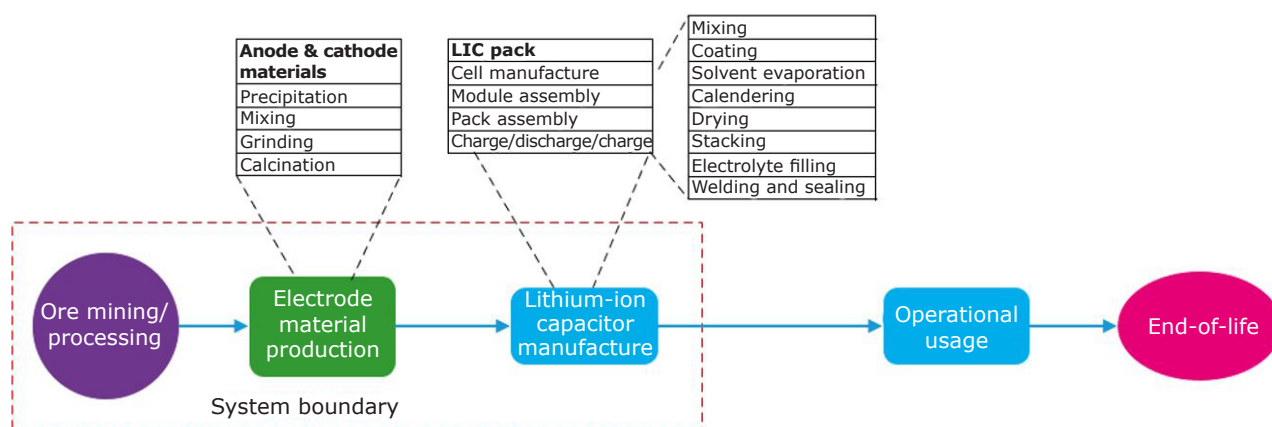


Fig. 1. LCA system boundary for the 48 V LIC manufacture from primary ore materials

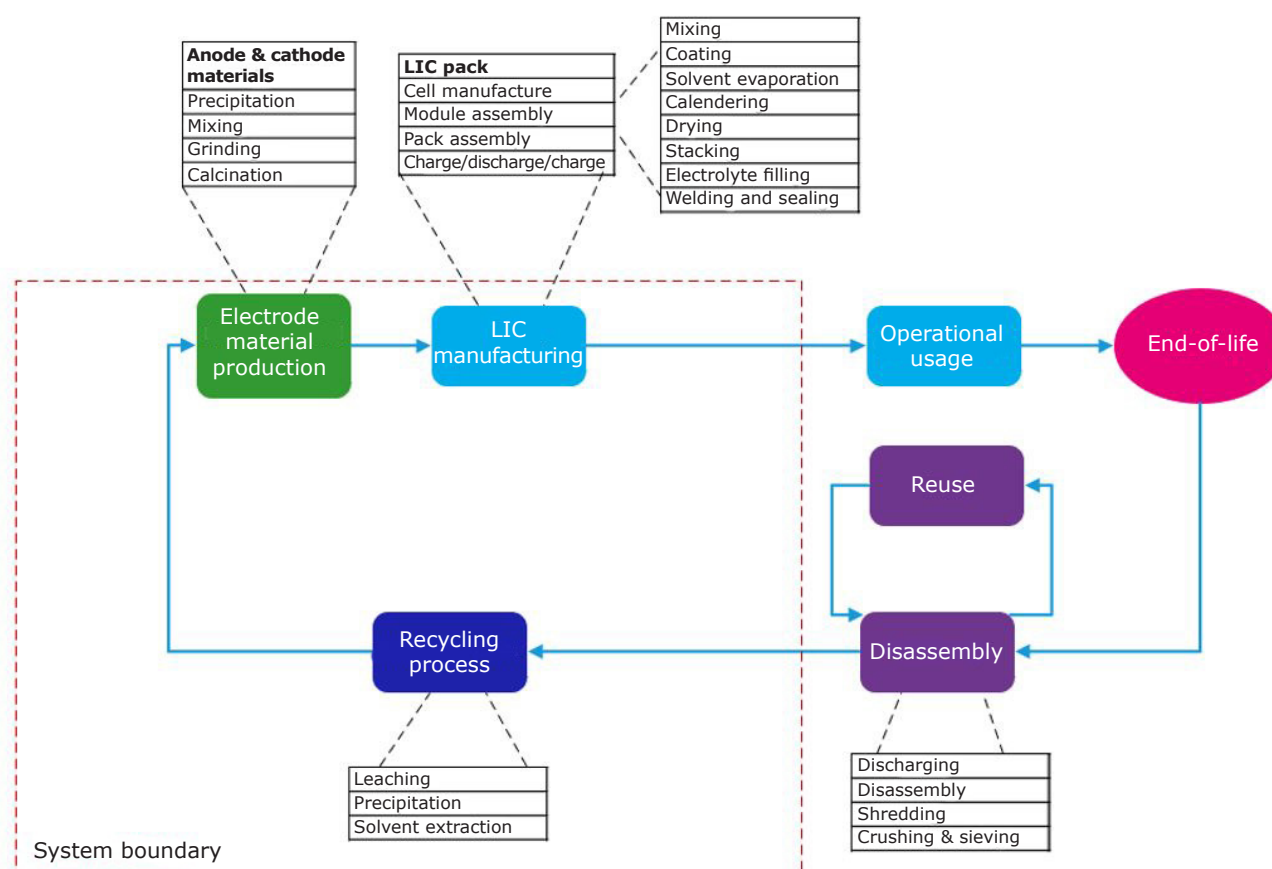


Fig. 2. LCA system boundary for the 48 V LIC manufacture from recycled end-of-life LIC

in **Figure 2** also excluded the disassembly and reuse process stages.

2.3 Methods and Databases

The commercial LCA software SimaPro 9.0 (PRé Sustainability, The Netherlands) was used in the study which utilised the ecoinvent 3.5 database

(ecoinvent, Switzerland). The Greenhouse gases, Regulated Emissions, and Energy use in Technologies (GREET®) 2017 model published by Argonne National Laboratory, USA, was also used when estimations of energy and reagent usage could not be determined from the commercial database. The relevant elementary flows of the starting material for the LIC manufacture using

recycled materials were obtained from a flowsheet model built in the gPROMS Process Builder software (PSE, UK). The ISO 14044 guidelines were only applied to the recycling flowsheet for the end-of-life LIC with application of the stepwise allocation procedure for multifunctional processes.

The LCA assumes raw materials were acquired from the market with global market average values used to evaluate the environmental burdens associated with the relevant material sourcing. This assumption did not apply to the LTO that was obtained from the recycling process. The emissions and energy associated with transportation were not considered in the study. However, the ecoinvent 3.5 database does account for the environmental impact associated with mining and transporting the various materials to the market. The electricity and heat energy sources used are for the UK with built-in ecoinvent database values used for environmental impact calculations.

3. Life Cycle Inventory

The elementary flows of material required to make the cells for a 48 V LIC module are based on pilot plant data. The recycled process data is based on a laboratory flowsheet that is simulated using a process model with appropriate scaling of model parameters from gram scale to a full-scale production plant. The main product and process stages for the primary (ore) and recycled materials manufacture of LIC are as follows: (a) anode powder material preparation;

(b) anode preparation; (c) cathode preparation; (d) electrolyte preparation; (e) cell formation; and (f) recycling of LTO powder (recycled material process only).

The detailed breakdown of materials for the assemblies and product stages of the two LCA comparative projects is in **Table I**. The only difference between the two comparative studies is in the source of lithium carbonate and titania for making the anode LTO powder material. For the primary (ore) process, the information for the environmental footprint associated with lithium carbonate and titania is obtained from the ecoinvent 3.5 database based on ore extraction and salt formation environmental impact values. However, in the case of the LIC module made from recycled materials, lithium carbonate and titania are obtained from the recycling product stage and only have process environmental impact values associated with reagents and energy consumption demand to recycle the end-of-life LTO anodes.

The supplementary data which contains the flowcharts and inventory to produce a lithium ion capacitor module and the list of assumptions used in the study is located with the online version of this article.

3.1 LTO Powder Synthesis

The information for the LTO powder used in the anode preparation process was not available in the ecoinvent 3.5 database or GREET® 2017. To determine elemental material flows of lithium

Table I Detailed Product Stages and Assemblies for the Primary Ore and Recycled Materials Lithium-Ion Capacitor Module

Primary (ore) LIC	Recycled materials LIC
–	Recycling 1.2 kg of LTO coated to anodes
Anode material: LTO powder preparation	Recycled material anode material: LTO powder preparation
Anode preparation: LTO slurry coating, dry and calender	Recycled material anode preparation: LTO slurry coating, dry and calender
Cathode preparation 1: slurry preparation	Cathode preparation 1: slurry preparation
Cathode preparation 2: coat, dry and calender	Cathode preparation 2: coat, dry and calender
Electrolyte preparation 1: dimethyl carbonate formation	Electrolyte preparation 1: dimethyl carbonate formation
Electrolyte preparation 2: Vinylene carbonate	Electrolyte preparation 2: Vinylene carbonate
Cell formation 1: cutting, stacking and drying	Cell formation 1: cutting, stacking and drying
Cell formation 2: electrolyte fill and packaging	Cell formation 2: electrolyte fill and packaging
Formation of 48 V LIC (primary ore)	Formation of 48 V LIC (recycled material)

carbonate and titania required to synthesise LTO an assumption of manufacture by solid-state reaction route was utilised (23). To account for lithium losses during the high temperature heating process, 5% excess lithium carbonate to stoichiometric requirements was added. The process energy requirements for synthesising LTO were obtained from GREET® 2017 by assuming similarity to those of manufacturing lithium manganese oxide (LMO).

The environmental footprint associated with lithium carbonate and titania was obtained from the ecoinvent database for the primary (ore) process. For the recycled materials LIC, environmental footprint attributable to lithium carbonate and titania were obtained as fractional contribution of the LTO anodes recycling product stage reagents and energy consumption.

3.2 Anode and Cathode Preparation

The elemental flows are for double side coating of 90 m of aluminium current collector foil from which 540 electrodes were made from the pilot plant. A 90% recovery and reuse assumption for *N*-methyl-2-pyrrolidone (NMP) was applied to the life cycle inventory as this is the expected design requirement at production scale. Without this assumption of NMP recovery and recycle the environmental impact from this organic solvent would be overestimated.

The cathode preparation stage elemental flows are based on the preparation of 180 electrodes from 30 m double sided coating on an aluminium foil from the pilot facility.

3.3 Electrolyte, Cell and Lithium-Ion Capacitor Module Formation

The LIC electrolyte consists of lithium hexafluorophosphate, ethylene carbonate, dimethyl carbonate and vinylene carbonate. Dimethyl carbonate and vinylene carbonate were not in the ecoinvent 3.5 database. These two components were assumed to have been synthesised from base materials using stoichiometric considerations.

The cells for the LIC are A5 pouch cells and each cell contained 11 anodes and 10 cathodes. The electrode cutting yield was 70% and the overall cell build yield value was 89% based on the pilot facility data. The failure rate of cells on testing was assumed to be 2% with the rejected cells discarded as waste. The scope of the study is for a 48 V LIC module and this was assembled from 160 cells.

3.4 Modelling of LTO Recycling Process

A proposed hydrometallurgical recycling flowsheet developed for recycling the LTO powder is shown in **Figure 3**. At the front end of the recycling process flowsheet, removal (decoating) of the LTO powder from the aluminium foil is executed by application of formic acid and this is then followed by a leaching stage using hydrochloric acid with a filtration stage which recovers titania. The filtrate undergoes a concentration step through evaporation followed by precipitation using sodium carbonate. Titania and lithium carbonate which are the main products from the recycling flowsheet are then used as starting feed materials for making the 'recycled material anode'.

The laboratory scale input values were used to inform a flowsheet model which was used to populate reagent and energy demand of the various processing stages. Appropriate scaling of model parameters from gram scale laboratory information to full scale production was applied in determining elementary flows of the recycled LTO materials.

4. Analysis of Life Cycle Assessment Results

While results for several environmental impact categories were available for analysis, for purposes of this study climate change (kilogram of CO₂ equivalent) and terrestrial acidification (kilogram of SO₂ equivalent) were analysed in greater detail for comparing the LIC module manufacture from primary ore materials against the recycled material process. The calculations are based on the ReCiPe Midpoint (H) with European Normalisation (24). The ReCiPe method was utilised because of its environmental relevance to the scope of the study, transparency and reproducibility. However, other methods which are also compatible with ISO standards could have been applied to the study.

Aluminium had the highest climate change and terrestrial acidification burdens to the extent of overshadowing contributions from other materials. To facilitate detailed analysis of environmental burdens of the other materials and processes, visual graphics of the results were plotted without the contribution from aluminium. Aluminium has established recycling processes but the decision if the quality of this recycled aluminium was of specifications sufficient for direct use in LIC manufacture was indeterminate and therefore

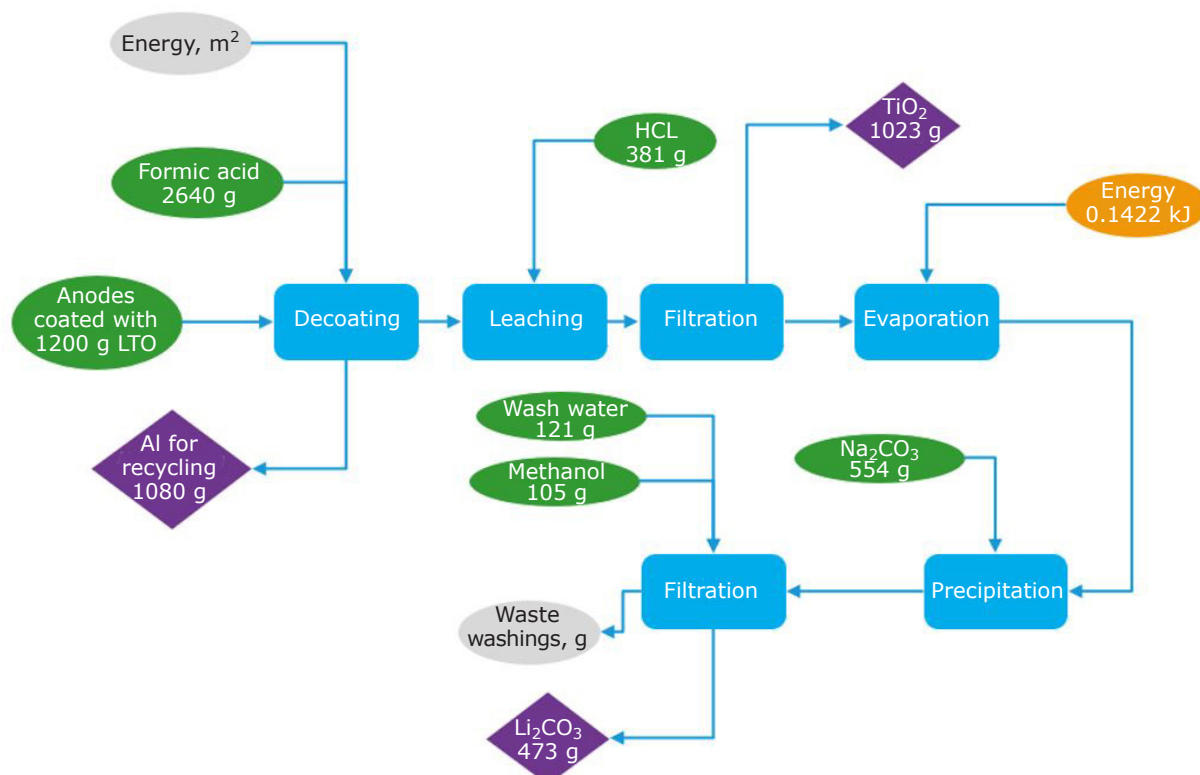


Fig. 3. Recycling flowsheet of anodes coated with 1.2 kg of LTO using metal recoveries from laboratory experiments

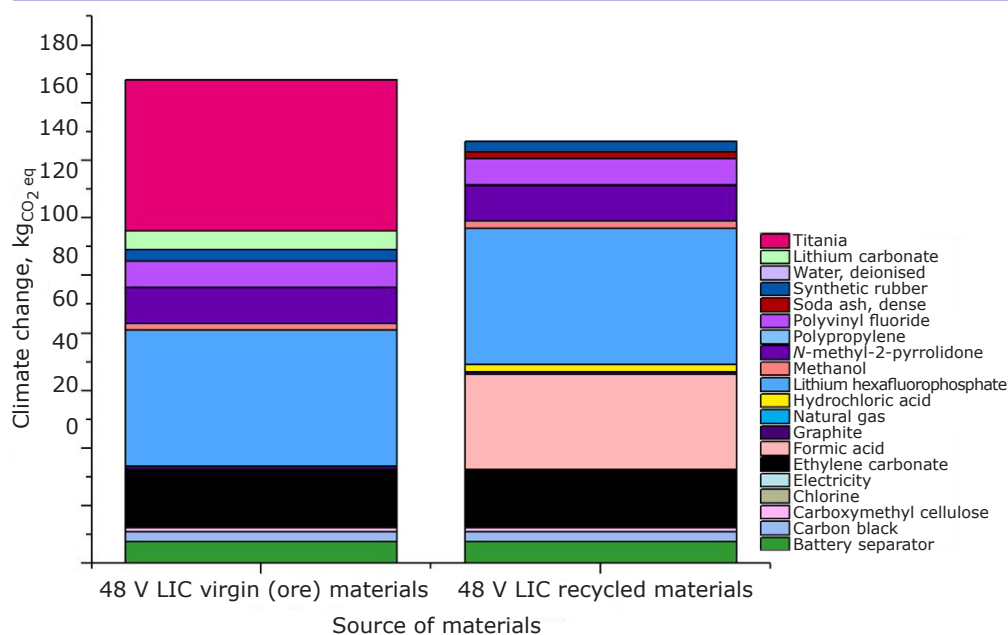


Fig. 4. Comparison of climate change associated with the production of a 48 V module LIC from primary ore materials and recycled LTO precursors (excluding aluminium contribution)

the LCA credit process was not applied towards aluminium used. **Figure 4** compares the climate change impact for making a 48 V LIC module using primary ore material and recycled LTO. Overall, utilising recycled LTO materials reduces the climate change impact by 12%. The order of decreasing

climate change for the LIC module manufacture using primary (ore) materials is titania > lithium hexafluorophosphate > ethylene carbonate. For LIC module manufacture using recycled LTO, the order of decreasing climate change is lithium hexafluorophosphate > formic acid > ethylene

Table II Comparison of Climate Change Contributions of the Main Product Stages for the Manufacture of a 48 V Lithium-Ion Capacitor Module Using 160 Cells from Primary (Ore) and Recycled LTO Materials^a

	Climate change, kg _{CO₂ eq}			
	Anode preparation	Cathode preparation	Cell formation	48 V LIC module (160 cells)
Virgin (ore)	82 (222)	8 (154)	78 (145)	168 (521)
Recycled	61 (198)	8 (154)	78 (145)	147 (497)

^a Values in brackets in the table include the contribution from aluminium

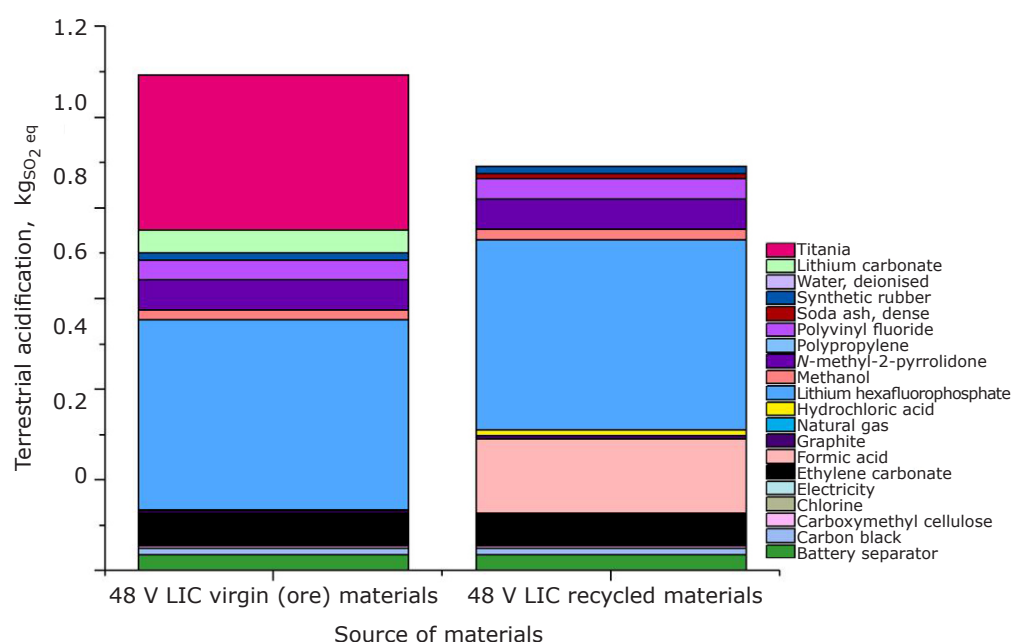


Fig. 5. Comparison of terrestrial acidification associated with the production of a 48 V module from primary ore materials and recycled LTO precursors (excluding aluminium contribution)

carbonate. The highest contributor towards climate change for primary (ore) case is titania while for the recycled LTO it is the lithium hexafluorophosphate electrolyte. Lithium hexafluorophosphate and ethylene carbonate are both part of the electrolyte system and have significant contributions which are equal for LIC manufacture using either primary (ore) or recycled LTO materials. Therefore, significant reductions in climate change for LIC manufacture using recycled LTO can only be achieved by reducing the quantities of formic acid used. **Table II** shows the climate change impact over the various stages of manufacturing a 48 V LIC module. The anode preparation stage has the highest contribution towards climate change for the two comparative cases. However, using recycled LTO lowers the climate change impact by 21 kg_{CO₂eq} compared to using primary (ore) during the anode preparation stage. The cathode preparation and cell formation stages have the same values as the two cases only differ in source of materials used the anode preparation stage.

Comparative analysis of terrestrial acidification for producing a 48 V LIC module using primary (ore) and recycled LTO materials is shown in **Figure 5**. Usage of recycled LTO for the anode manufacture product stage results in 18% reduction in terrestrial acidification compared to using primary ore materials. The major contributors towards terrestrial acidification in decreasing order are lithium hexafluorophosphate > titania > ethylene carbonate for LIC module manufacture using primary (ore) materials. For LIC manufacture using recycled LTO, the major contributors towards terrestrial acidification in decreasing order are lithium hexafluorophosphate > formic acid > ethylene carbonate. **Table III** shows the terrestrial acidification associated with the various stages of manufacturing a 48 V LIC module for the two cases. The anode preparation stage has the highest contribution towards terrestrial acidification when the primary (ore) and recycled LTO material sources are compared. By utilising recycled LTO for the anode preparation

Table III Comparison of Terrestrial Acidification Impact of the Main Product Stages for the Manufacture of a 48 V Lithium-Ion Capacitor Module Using 160 Cells from Primary Ore vs. Recycled LTO Precursors^a

	Terrestrial acidification, kg _{SO₂eq}			
	Anode preparation	Cathode preparation	Cell formation	48 V LIC (160 cells)
Virgin (ore)	0.51 (1.27)	0.04 (0.83)	0.55 (0.90)	1.1 (3.00)
Recycled	0.30 (1.06)	0.04 (0.83)	0.55 (0.90)	0.89 (2.79)

^a Values in brackets in the table include the contribution from aluminium

process the terrestrial acidification impact is lowered by 0.21 kg_{SO₂eq} compared to using primary (ore) materials.

While there are environmental benefits from using recycled LTO, the existing recycling process flowsheet has a lot of optimisation opportunities especially regarding the quantities of formic acid used which have a significant contribution towards both climate and terrestrial acidification.

5. Application of Sustainability and LCA in Early Phase R&D Activities

The application in early phase R&D activities is demonstrated in this section as applied to process development choices for the recycling stage. Before the hydrometallurgical treatment detailed in Section 3.4, the LTO has to be decoated (removed) from the aluminium foil to which it is bound. The widely used binder for coating the LTO and most active materials to current collectors is polyvinylidene fluoride (PVDF) because of its adhesive capabilities and electrochemical stability (25). The PVDF binder presents a challenge to the decoating process as it is only partially soluble in most common solvents. The common solvent for dissolving PVDF is NMP which is also used during the slurry coating process. However, NMP has high environmental and toxicity burdens which has resulted in stringent legislative restrictions of its usage (26).

To improve the sustainability metrics of the recycling process, several alternative solvents were investigated for their capabilities to remove the LTO from the aluminium foils. Acetone and polyethylene glycol (PEG) have similar properties to dipolar aprotic solvents like NMP and were identified as greener alternatives (27). Several other organic reagents such as acetic acid, formic acid, ethylene glycol and methanol were also screened as potential candidates for the process. **Figure 6** shows images of LTO anodes after stirring in different reagents for 1 h at room temperature. From **Figure 6**, formic acid has higher technical performance compared to the other solvents as it removed all of the visible traces of carbon and LTO from the aluminium foil.

The decision-making process considered the environmental footprint of these reagents in addition to their technical performance for removing the LTO from the aluminium foils. **Figures 7(a)** and **7(b)** show the contribution towards climate change and terrestrial acidification respectively based on using 1 kg of these reagents. NMP has the highest environmental burden for climate change and terrestrial acidification, while methanol has the least environmental footprint. However, methanol efficiency in decoating LTO was low and therefore a trade-off of technical performance, environmental impact and costs resulted in formic acid as the alternative reagent choice for the decoating process stage. The bulk purchase price for these reagents is in **Table IV**.

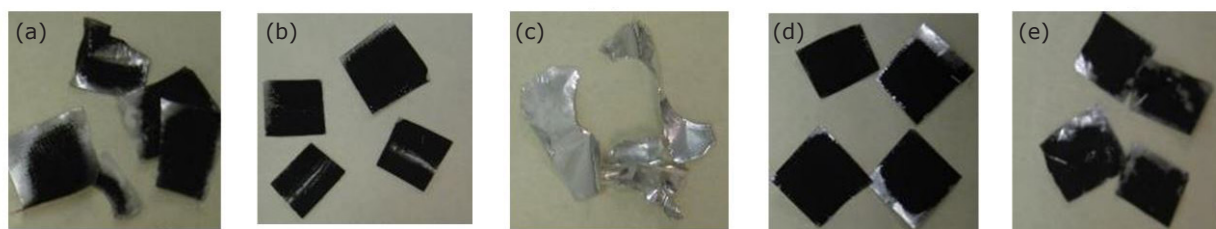


Fig. 6. LIC anode foils after stirring in solvent for 1 h at room temperature: (a) acetone; (b) acetic acid; (c) formic acid; (d) methanol; (e) PEG

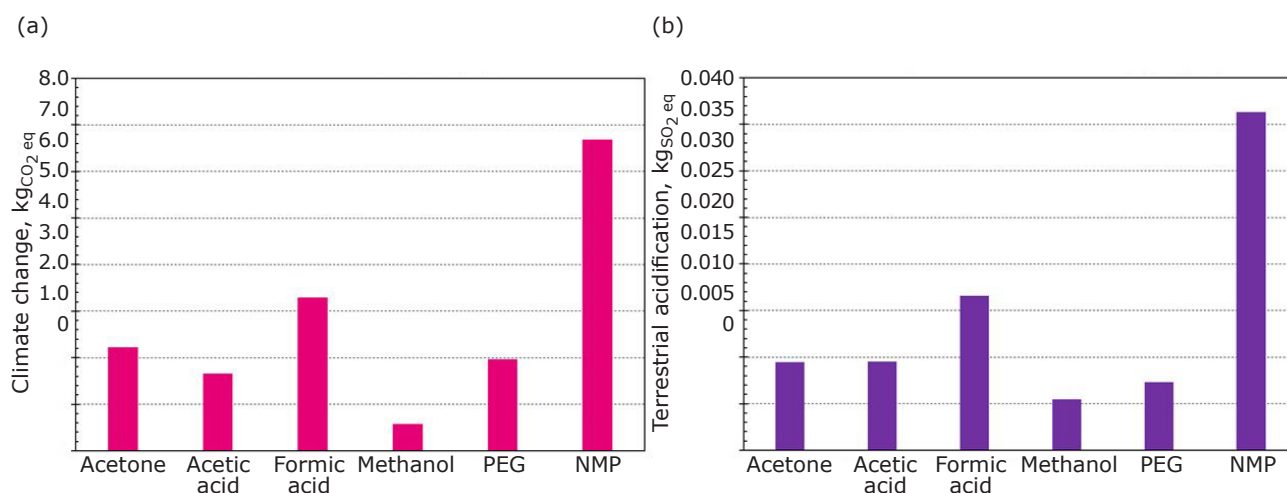


Fig. 7. Environmental impact: (a) climate change; and (b) terrestrial acidification of potential reagents trialled for separating LTO from the aluminium foil current collector on the basis 1 kg usage of the reagents

Table IV Bulk Chemicals Purchase Price of Potential Reagents Trialled for Decoating LTO^a

	Decoating reagent				
	Acetone	Acetic acid	Formic acid	Methanol	NMP
Price, £ l ⁻¹	5.8	60.4	66.6	23.4	95.5

^a ICIS October 2019 prices (28)

While several product and process research activities are focussing on novel binders that are less toxic and low costs compared to PVDF (29), the electrodes bound with PVDF that have already been manufactured will still require a more environmentally sustainable process to recover the active material at their end-of-life. The approach applied in this case study of early phase R&D process development activities demonstrated sustainable choices in alternate reagent selection in alignment to the triple bottom line approach (30). The choice of formic acid when compared to NMP results in intersection of people, planet and profit (3Ps) requirements of sustainability. For this case study, formic acid had reduced environmental impact, toxicity and meets the profit criteria through high decoating technical efficiency at lower costs compared to NMP.

6. Conclusions

The LCA methodology was applied to quantitatively determine the environmental burdens associated with manufacturing a 48 V LIC module. The prospective LCA compared the environmental impact of manufacturing a LIC module using primary ore materials versus LIC manufacture using recycled materials from end-of-life LICs. The

anode preparation stage is associated with most of the environmental burden for manufacturing the LIC module for both processes due to the source of precursors used in production of the active LTO material. Utilisation of LTO precursors from recycled end-of-life LICs reduced both climate change and terrestrial acidification environmental impact categories for the LIC module manufacture. However, the sustainability metrics of the recycled process route of production could potentially be improved further by optimised application of formic acid which is used in the process stage for separating the LTO from the aluminium current collector foils.

The application of the LCA methodology in early phase R&D activities was demonstrated for the process development reagent choice case study. The LTO decoating reagent decision-making process considered the environmental footprint, technical performance and costs. The decision to utilise formic acid as a decoating agent was a sustainable choice which balanced environmental, economic and social performance. For the demonstrated case study, the choice of formic acid as decoating reagent reduced climate change and terrestrial acidification, lowered human toxicity values and met the profit criteria through high separation efficiency at lower costs.

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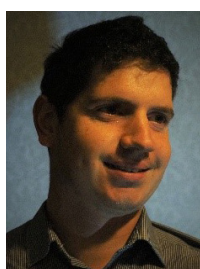
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Alexander Roberts worked as a principal engineer at WMG. He is now an Associate Professor in Energy Storage at Coventry University, UK, and a Faraday Institution Industrial Fellow. He leads activities in development and prototyping of energy storage technologies, including lithium- and sodium-ion batteries, supercapacitors and hybrid devices.

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SUPPLEMENTARY INFORMATION FOR:

**Comparative life cycle assessment of lithium-ion capacitors
production from primary ore and recycled minerals**

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<Summary>

The supplementary information contains:

S1 Flowcharts and inventory to produce a lithium ion capacitor module and

S2 A list of assumptions applied in the study.

S1.1 LTO Powder synthesis

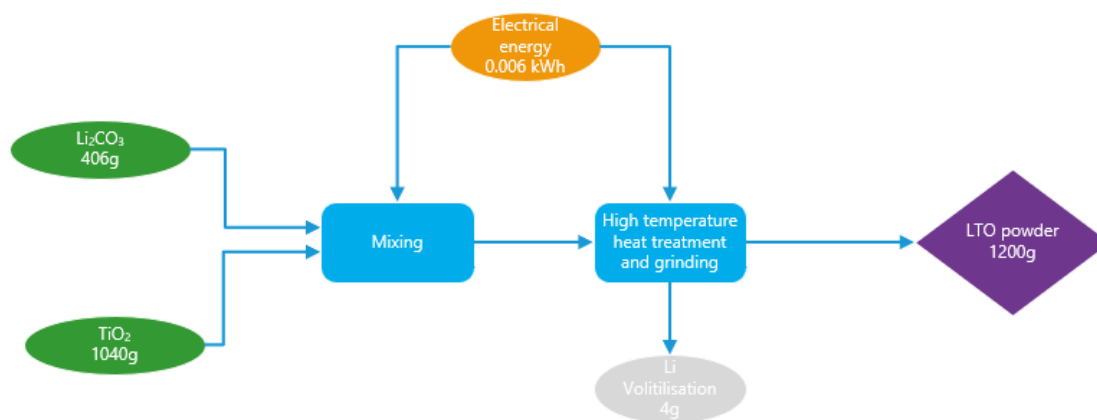


Figure S1. Anode powder material synthesis flowsheet and inventory

S1.2 Anode preparation

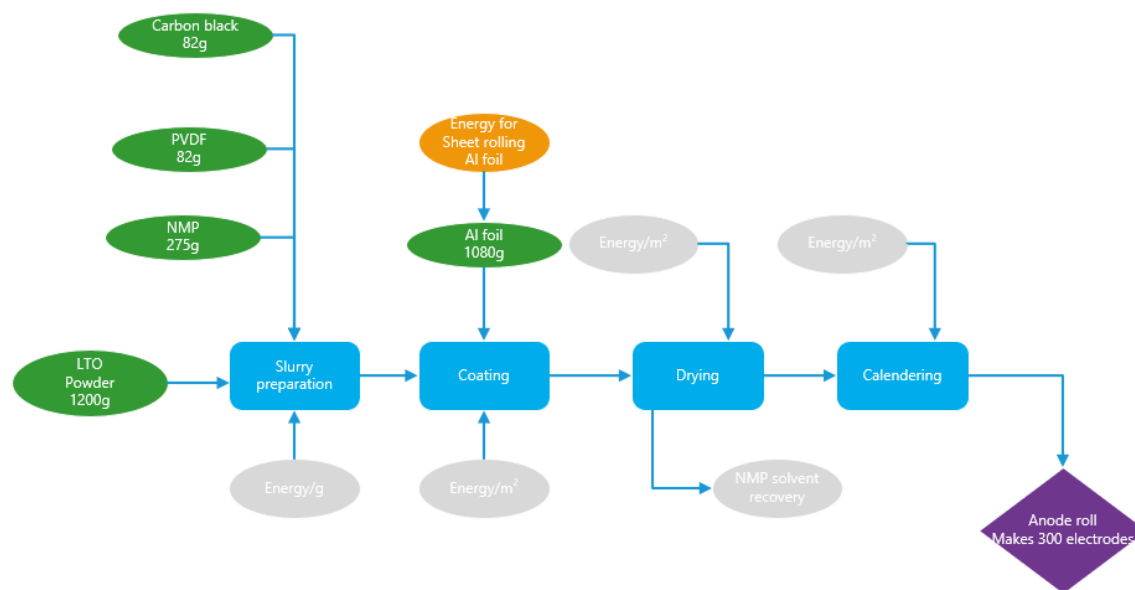


Figure S2. Anode preparation assemblies and product stages

S1.3 Cathode preparation

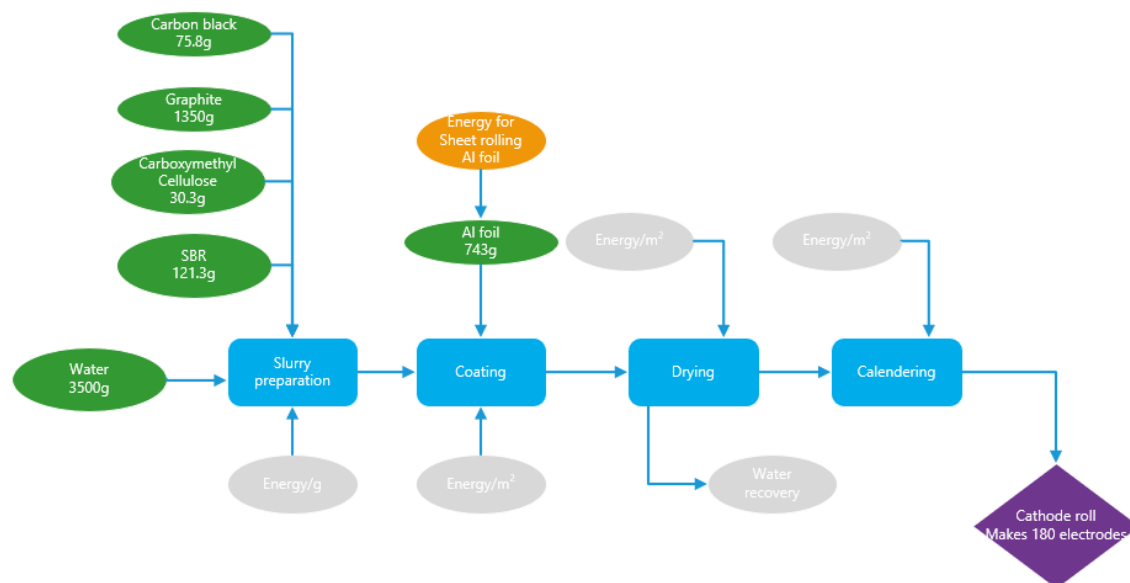


Figure S3. Cathode preparation assemblies and product stages

S1.4 Cell build

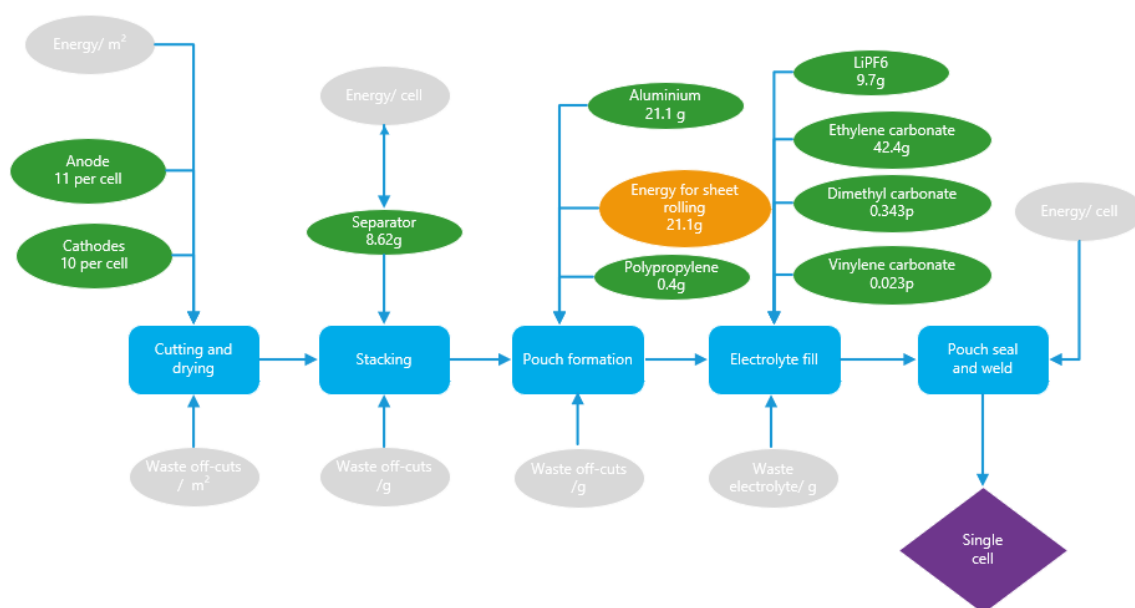


Figure S4. Flowsheet for a single cell formation

S2.0 LCA assumptions for the 48V LIC module**Table S1**

Process stage	Data source	Assumption
All	–	Emissions and energy associated with transport has not been considered in this LCA. But the Ecoinvent database does include transport associated with primary ore mining of minerals.
Anode Material: LTO synthesis	Zhang <i>et al.</i> 2013	Energy requirements for LTO synthesis are similar to LMO with data obtained from Greet model
Anode preparation: anode slurry coating	Pilot plant	90 % recovery and reuse of NMP assumed at plant production scale. Polyvinylidene fluoride (PVDF) absent in Ecoinvent database and polyvinyl fluoride (PVF) used as surrogate.
Cell formation: electrolyte & housing	–	Diethyl carbonate absent in Ecoinvent database, stoichiometric consideration applied to make diethyl carbonate from ethylene carbonate and methanol
Cell formation: electrolyte & housing	–	Vinylene carbonate absent in Ecoinvent database, stoichiometric consideration applied to make vinylene carbonate from ethylene carbonate and chlorine
Cell build	Pilot plant	Cell build yield of 89%
48 V LIC module	Pilot plant	2% failure rate of cells
Recycling end-of-life LIC	Laboratory flowsheet	Estimate of energy for LTO decoating, leaching and precipitation based on scale-up values using process model

Reference

Q. Zhang, C. Zhang, B. Li, S. Kang, X. Li and Y. Wang, *Electrochim. Acta*, 2013, **98**, 146. doi: 10.1016/j.electacta.2013.03.006