Prospective LCA of the production and EoL recycling of a novel type of Li-ion battery for electric vehicles

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Abstract

The transport sector as a whole – and within it passenger cars in particular – is currently responsible for a large share of the total greenhouse gas emissions of many developed and developing countries, and a transition to electric vehicles (EVs) is often seen as a key stepping stone towards the de-carbonization of personal mobility. Research is on-going in the continuous development and improvement of lithium ion (Li-ion) batteries, which may use a range of several different metals in conjunction with lithium itself, such as: lithium manganese oxide (LMO), lithium iron phosphate (LFP), lithium nickel cobalt manganese oxide (NCM), and lithium nickel-cobalt-aluminium oxide (NCA). Within the MARS-EV research project, a new cell chemistry has been developed and tested, using a lithium cobalt phosphate (LCP) formulation. This work presents the first life cycle assessment (LCA) for

such LCP batteries, including a newly-developed hydrometallurgical battery recycling process which enables the end-of-life recovery of not only the valuable metals, but also of the graphite component, thereby avoiding the associated CO_2 emissions.

Keywords: LCA; Li-ion; battery; electric vehicles; end of life.

1. Introduction

The automotive sector is gradually embracing electric power trains in replacement of the internal combustion engines (ICEs) that have heretofore been the *de facto* standard. In the UK, for instance, electric vehicle (EV) sales are growing exponentially (Nextgreencar, 2017), and the government has pledged to end all sales of conventional ICE vehicles by 2040 (UK Gov, 2017). This move is intended primarily as a means to significantly curb the levels of airborne pollutants in densely populated areas such as cities and towns, with the additional side benefits of potentially reducing the vehicles' life-cycle cumulative energy demand and greenhouse gas emissions too. However, while the reductions in polluting emissions during the use phase are automatically achieved by all electric vehicles (EVs), the latter's comparative environmental and energy performance on the full life cycle scale *vs*. that of conventional ICE vehicles ultimately depends on two critical factors: (i) the way in which the electricity that powers the vehicles is generated, and (ii) the indirect energy and emissions that are 'embodied' in the large and heavy battery packs that are used to store that energy.

With regards to (i), the scientific literature is in broad agreement in identifying ample scope for solid environmental benefits in the use phase of EVs, when compared with conventional ICE vehicles. For instance, Faria et al. (2013) found that, depending on the grid supply mix of the country where they are driven, greenhouse gas (GHG) emissions by compact EVs may be reduced by as much as 60% with respect to those of ICE cars of the same size segment. Similarly, Raugei et al. (2018) found that the use-phase demand for non-renewable primary energy of a compact EV in the UK is currently lower by almost 60% with respect to that of an

otherwise similar ICEV, and that such result may be expected to improve even further in a range of future grid mix scenarios.

The indirect impacts due to the battery packs (ii) are instead less straightforward to pin down. A wide range of results are presented in the scientific literature, since a large share of such impacts is due to the materials used in the battery cells and the related production processes, and these tend to vary significantly depending on the specific chemistries involved and on the vintage of the data.

Lithium-ion battery (LIB) cells are fundamentally composed of a cathode, an anode, an electrolyte (to enable closure of the electrical circuit by ionic transport) and a separator (to prevent the anode and cathode from touching and thus short-circuiting the cell). The anode, electrolyte and separator are relatively invariant across most LIBs, and specifically: the anode is usually made of carbon (graphite), bound to a copper conductor plate using a polymer binder; the electrolyte is an organic liquid with dissolved ionic salts (such as $LiPF_6$); the separator is made of a porous plastic polymer. Hence, it is the composition of the cathode that is most responsible for setting apart the various battery types, and the choice of cathode entails a number of trade-offs in terms of key performance criteria, besides energy density.

The most commonly employed and analysed cathode types to date are: lithium manganese oxide (LMO = $LiMn_2O_4$), lithium iron phosphate (LFP = $LiFePO_4$), lithium nickel-cobalt-manganese oxide (NMC = $LiNi_xCo_yMn_zO_2$), and lithium nickel-cobalt-aluminium oxide (NCA = $LiNiCoAlO_2$) (Zubi et al., 2018).

LMO and LFP batteries may generally be considered the least environmentally critical, since they do not contain particularly toxic or rare metals. They are also relatively mature technologies, which have been available commercially since the mid 1990s. Both were used in early EVs, especially LFP which can withstand a larger number of charge cycles and tolerate a wider state-of-charge range. However, both technologies are relatively inferior to

the cobalt-containing alternatives in terms of energy density, and have therefore been largely superseded by the latter in most modern EV applications.

NMC is at the moment the most widely-adopted LIB technology in current-production EVs, and also likely the most promising for the near future. It is comparatively very reliable, durable, and provides an improved balance between energy density and durability. NCA is also a viable alternative technology for those EV applications in which an even greater energy density is required, if at the same time a slightly reduced durability is acceptable.

In this paper, we present a prospective life cycle assessment (LCA) of the production and end-of-life (EoL) recycling of a new, high-energy type of cobalt-containing LIB for EVs, using a lithium cobalt phosphate (LCP = $LiCoPO_4$) cathode, which was developed within the MARS-EV research project. While other research efforts targeting similar LCP cathode chemistries are documented in the recent literature (Örnek, 2017; Mauger et al., 2017; Ke et al., 2017; Fang et al., 2016), never before has a LIB utilizing such a cathode been the object of an LCA.

Then, we alsodiscuss our findings within the context of a range of previously published results for LIBs using more conventional chemistries.

2. Materials and methods

LCA is a tool that has rapidly grown to become standard procedure for scientists and engineers to investigate and assess the environmental performance of a wide range of human-dominated processes. In the late 1990's the International Standardization Office started a large effort towards the comprehensive standardization of Life Cycle Assessment, which culminated in the current set of standards (ISO 2006a,b).

This section contains a detailed discussion of the three main stages into which our assessment of the production and end-of-life (EoL) recycling of a novel type of Li-ion battery for electric vehicles is articulated.

2.1 Goal and scope definition

The goal of our analysis was two-fold: firstly, to assess the total demand for primary energy and the cumulative greenhouse gas (GHG) emissions of a new type of Li-ion battery which was developed within the EU FP-7 'MARS-EV' research project; and secondly, to compare the results obtained for this type of battery to those for more conventional Li-ion battery alternatives.

In terms of scoping, as illustrated in Figure 1, the analysed system consists of two sections:

- all the manufacturing steps that lead to a complete 17kWh battery pack (total mass = 108 kg) to be used in compact EVs ('cradle-to-gate' boundary);

- a novel EoL treatment aimed at achieving high recycling rates and the corresponding energy and carbon emission credits ('cradle-to-gate' + End-of-Life boundary).

Given that the focus of this study is specifically on the new chemical formulation of these batteries and on how these compare to existing alternatives, the use phase (which strongly depends on the assumed driving cycle and on the grid mix that supplies the electricity for re-charging, but is substantially independent from the chemistry of the batteries themselves) has been deliberately omitted.

From a practical standpoint, the entire LCA was carried out making use of the latest version of the GaBi Professional LCA software package (Thinkstep, 2017).

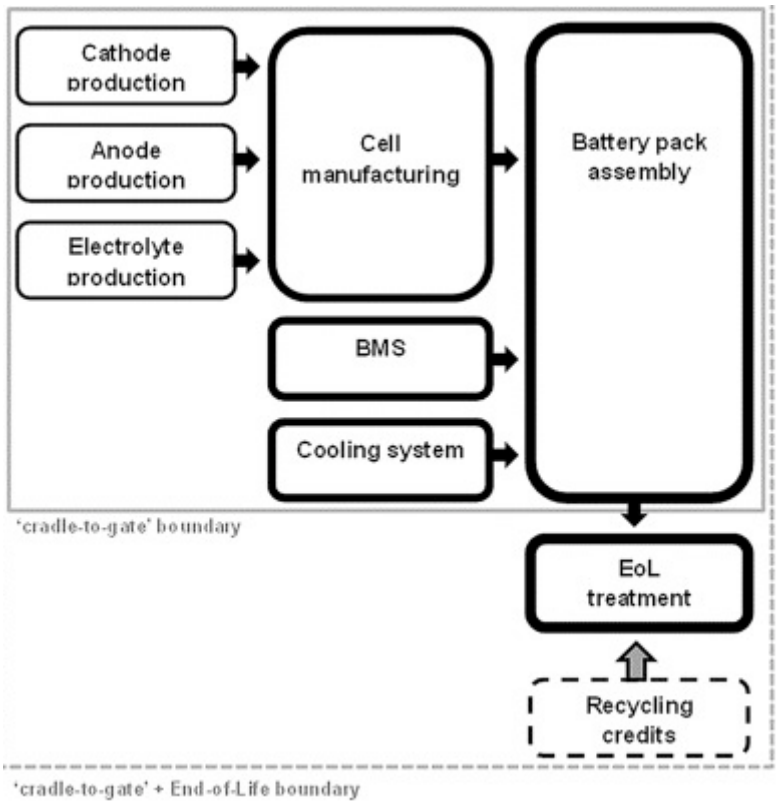

Figure 1 – Flow diagram of the main steps in the manufacturing of a complete Li-ion battery pack, with indication of separate assessment of end-of-life (EoL) treatment and associated recycling credits.

2.2 Inventory analysis

When compiling the life cycle inventory for the analysed system, the quality and reliability of the employed data is of paramount importance, since the results of the analysis cannot obviously be of any better quality than the input data themselves.

Three types of data sourcing are possible, in descending order of reliability and preferability:

1. Data obtained through direct measurements, made either by the LCA analyst him/herself or by the manufacturers of the analysed products. These are usually the most dependable data of all, however they may be very specific to the process under study, and should not be carelessly transferred to other analyses.

2. Data from previous studies available in the scientific literature, and more specifically, in up-to-date and cross-checked international LCI databases. The analyst has less direct control over these, but they have the advantage of having been reviewed by expert analysts before, and are often a weighted average of several similar processes.

3. Data inferred by means of educated guesses and estimates, based on previous analyses and the analyst's experience. These are inevitably affected by the greatest uncertainty; yet, if the analyst has sufficient experience in the field, they can often still be acceptable.

Additionally, LCA classifies input data as respectively pertaining to "foreground" or "background" processes. The former are those "…that are under direct control of the producer of the good or operator of the service, or user of the good or where he has decisive influence…This covers firstly all in-house processes of the producer or service operator of the analyzed system. Secondly…also all processes and suppliers of purchased made-to-order goods and services, i.e. as far as the producer of service operator of the analyzed system can influence them by choice or specification". (JRC, 2010, p.97) Conversely, the latter are "…those processes that are operated as part of the system, but that are not under direct control or decisive influence of the producer of the good (or operator of the service, or user of the good). The background processes and systems are hence outside the direct influence or choice of the producer or service operator of the analyzed system." (JRC, 2010, p. 98).

In our analysis, we relied to the maximum extent possible on Type-1 data (directly supplied by the MARS-EV project partners) for the manufacturing of all cell components, as well as for the direct inputs and outputs of the EoL treatment processes.

This information was complemented with Type-2 data for those parts of the system which were not directly resulting from the work done by the MARS-EV consortium members, and specifically for:

(i) the “foreground” inputs to the manufacturing of the battery pack, including the outer packaging, the electronics (battery management system) and the cooling system;

(ii) all “background” inputs for the supply chains of the direct material and energy inputs used in the manufacturing and EoL processes.

The main sources used for these Type-2 data were the latest versions of the BatPac software (Argonne National Laboratory, 2015), and of the reputable Ecoinvent LCA database (Ecoinvent, 2016). More specifically, for the purposes of producing results which could be considered representative of the expected average energy and carbon performance of the novel battery type under analysis, assuming that the latter would be commercially produced and deployed in Europe, all electricity inputs for all production stages were modelled according to the current average ENTSOE (formerly UCTE) European grid mix.

Finally, the use of Type-3 data (educated guesses and assumptions) was limited to two instances related to the manufacturing of the lithium cobalt phosphate, as explained in more detail in Section 2.2.1.

2.2.1 Battery cells

The newly developed MARS-EV LIB utilizes lithium cobalt phosphate (LCP) as the key electroactive material for the cathode. LCP is a high-energy cathode material selected in the project for synthesis by flame spray pyrolysis (conducted by Johnson Matthey, JM, UK). As a result of the high de-/lithiation potential at about 4.8 V *vs*. Li/Li+, LCP provides a theoretical specific energy of ca. 850 Wh/kg, i.e., a value very close to the targeted 900 Wh/kg at cathode level to be able to develop a high energy density Li-ion battery cell (>250 Wh/kg). Best processability using environmentally friendly water-based slurries for electrode production as well as better electrochemical performance were obtained when applying a carbonaceous coating to the LCP particles. Thus initial reversible capacity of 120 mAh/g and still 70 mAh/g after 112 cycles were achieved (compared to ca. 20 mAh/g for the non-coated LCP-based electrodes). Accordingly, the large-scale synthesis was modified to provide in situ

carbon-coated spherical LCP secondary particles, consisting of fine primary nanoparticles (patent application GB1609016.9).

Given the novelty of this material, the only information available on its production process inevitably referred to a pilot-scale batch. While this was considered to be still adequately representative of the intrinsic demands for the main technology-specific material inputs (including the key lithium and cobalt precursors), the reported values for the compressed oxygen and, importantly, electricity inputs appeared to be much larger than the corresponding inputs for the production of the more common chemistries as reported in the literature (Ecoinvent, 2016). This raised an issue in terms of the subsequent comparability of the results of this analysis to those from previous studies that refer to commercial scale production (*cf*. Section 3.2). Conservative estimates equal to twice the oxygen and electricity input values that are reported in the Ecoinvent LCA database for LMO production were therefore adopted here as probable better proxies for LCP production too, assuming further expansion of the latter to full commercial scale in the near future.

The electrochemistry of the MARS-EV battery cells is then completed by a comparatively conventional graphite anode. The inventory for the production of this component was compiled on the basis of a combination of Type-1 and Type-2 data.

The electrolyte selected for employment in the MARS-EV battery cells was a lithium-rich electrolyte ($LiPF_6$) in a blend of Ethylene Carbonate and Dimethyl Carbonate, plus organic additives. A severe challenge when dealing with such high-voltage cathodes, commonly cycled up to about 5.0 V vs. Li/Li+, concerns the anodic stability of the electrolyte. Improvement of the cycling stability of LCP electrodes and full cells was eventually realized by optimizing the electrolyte composition with adequate selection of the organic additives. The final electrolyte composition was supplied by MARS-EV project partners, and all the necessary precursors were inferred on the basis of standard organic chemistry stoichiometry.

Finally, the individual battery cells are isolated using a novel cellulose-based packaging material which is both lighter and less energy-intensive to produce than the more

conventional metallic alternatives. The “foreground” inventory for the production of this new packaging was also provided by MARS-EV project partners. The remaining quantitative information required to complete the cell manufacturing model was sourced from the BatPaC software.

2.2.2 Battery pack

According to the information contained in the BatPaC model (and revised and adapted in collaboration with all MARS-EV project partners), ninety-six battery cells were assumed to be assembled into four modules that together comprise a 17kWh battery pack. The latter is then complemented by the necessary electronics (the ‘battery management system’ – BMS) and a suitable cooling system. The bill of materials for all the manufacturing steps beyond the battery cell stage was informed by the BatPaC software.

2.2.3 End of Life

LIB battery recycling may still be regarded as being in its infancy, with only few medium-to-large scale commercially operated activities worldwide. Also, research is still under way to explore a number or alternative recycling processes and there is no consensus yet on which one will ultimately emerge as the most profitable and environmentally advantageous.

In general terms, all recycling process chains tend to be structured in a broadly similar way, as described below (Zeng et al. (2014):

1. Manual electrical control, discharge and dismantling.

2. Dry mechanical shredding and grinding (not strictly required for pyrometallurgical secondary treatment routes).

3. Secondary treatment. This is where the actual recycling takes place, which may happen via a range of pyrometallurgical (Glencore, 2017; Sumito, 2017; Umicore, 2017) or hydrometallurgical (Hanisch, 2014; Chen and Ho, 2018) processes. The latter tend to be less energy demanding, primarily because of the lower temperatures involved (Gaines et al., 2012).

After initial manual dismantling and dry mechanical shredding, the new EoL treatment process developed within the MARS-EV project relies on hydrometallurgical recycling, which entails an acid leaching and oxidizing step, leading to the recovery of a pure carbon cake (90% recovery rate), followed by the alkalinisation of the solution, which leads to the controlled precipitation and selective separation of the metal salts (90% recovery rates for aluminium, copper, lithium and cobalt).

Further details of these critical secondary treatment processes cannot be disclosed due to confidentiality agreements, but all necessary inputs quantities and associated emissions were either directly supplied by the responsible project partner or otherwise back-calculated by us and then checked and approved by them.

The key advantages of this new EoL treatment are two-fold: (i) a low energy consumption, leading to an overall demand for 148 MJ of primary energy (PE) per kWh of battery energy capacity sent to recycling, or 23 MJ(PE)/kg; and (ii) the ability to recover not only the valuable metals, but also the graphite component, thereby avoiding the associated "foreground" CO_2 emissions.

From a methodological perspective, energy and emission credits were assigned to the recovered metals according to a substitution logic, i.e.:

- The Al and Cu recovered in steps 1 and 2 were assumed to displace the respective current primary/secondary market mixes for those metals (Aluminium Federation, 2017; Copper Alliance, 2017);
- All the metals recovered in step 3 were assumed to displace the most common respective salts, with all stoichiometries duly adjusted.

The adoption of this strict substitution logic leads to lower calculated environmental credits compared to the alternative assumption that all the recovered metals would simply displace equivalent quantities of their virgin counterparts, as is sometimes done in other studies in the literature (Dunn et al., 2012; Hendrickson et al., 2015). However, the former methodological approach is arguably the more scientifically sound of the two, since the actual environmental

benefit of recycling is ultimately determined by what the recovered materials actually go on to displace in the market, and not by any optimistically assumed equivalence only based on their metal content (Ayres, 1988; Bala Gala et al., 2015).

2.3 Impact assessment

In this LCA, we decided to focus on Cumulative Energy Demand (CED) and Global Warming Potential (GWP) as the impact indicators of choice, because of (i) their overwhelming relevance, especially for energy using products (EuPs) such as batteries, and (ii) our intention to then compare our results to those of previous analyses (these two indicators are by far the most represented in the literature). In particular, it bears reminding that CED measures the total amount of energy that is cumulatively harvested from the full range of available primary resources (including fossil fuels, nuclear resources, and renewable resources such as biomass, solar, wind and hydro); a comprehensive explanation and discussion of the merits and limitations of this approach is available in the referenced literature (Frischknecht et al., 2007; 2015).

Finally, we deliberately refrained from applying any normalization and weighting to our results because, while potentially facilitating the interpretation of the results by a less technical audience, they inevitably remain the most arbitrary steps in any LCA, since the choice of the weighting factors is to a large extent political, with very little if any scientific relevance. In fact, because of this, according to ISO, normalization and weighting are always optional steps and are discouraged for any “comparative assertion intended to be disclosed to the public” (ISO, 2006b).

3. Results and discussion

3.1 Results for the MARS-EV battery pack

Figure 2 illustrates the cumulative energy demand of the analysed battery pack (expressed in terms of MJ of total primary energy input per kWh of battery energy capacity).

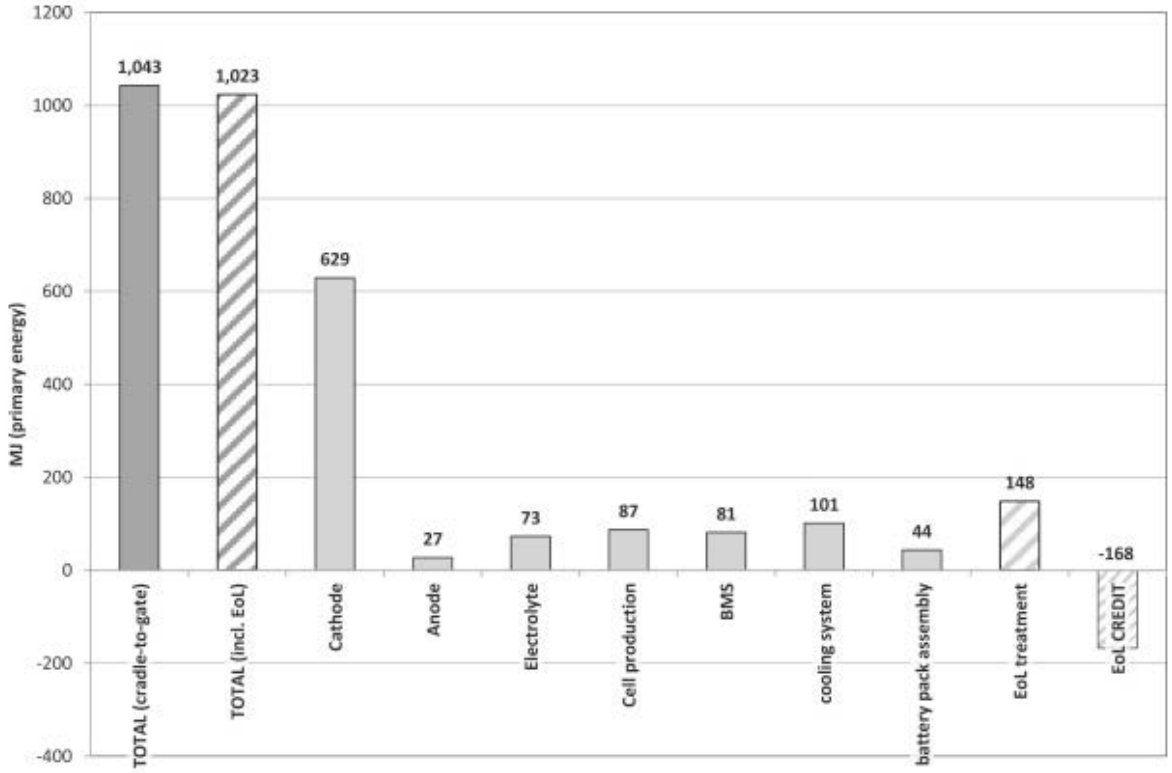

Figure 2 – Cumulative energy demand (CED) per kWh of battery energy capacity, with and without end-of-life (EoL), and highlighting the individual contributions of the main components (BMS = battery management system).

As clearly visible, the largest contributor by far (> 60%) to the CED of the entire battery pack was found to be the cathode. Interestingly, this result is largely determined by the 'embodied' energy in the input materials for the cathode (the latter being informed by direct pilot-scale measurements, and only marginally susceptible to further reduction), whereas the direct manufacturing energy input (which was estimated on the basis of literature information on large-scale LMO cathode production) is comparatively unimportant.

Also noteworthy is the fact that the inclusion of the EoL treatment steps and associated credits only leads to a marginal reduction of the total CED (-2%).

Figure 3 then illustrates the cumulative greenhouse gas (GHG) emissions of the analysed battery pack (expressed in terms of kg of CO_2-equivalents per kWh of battery energy capacity).

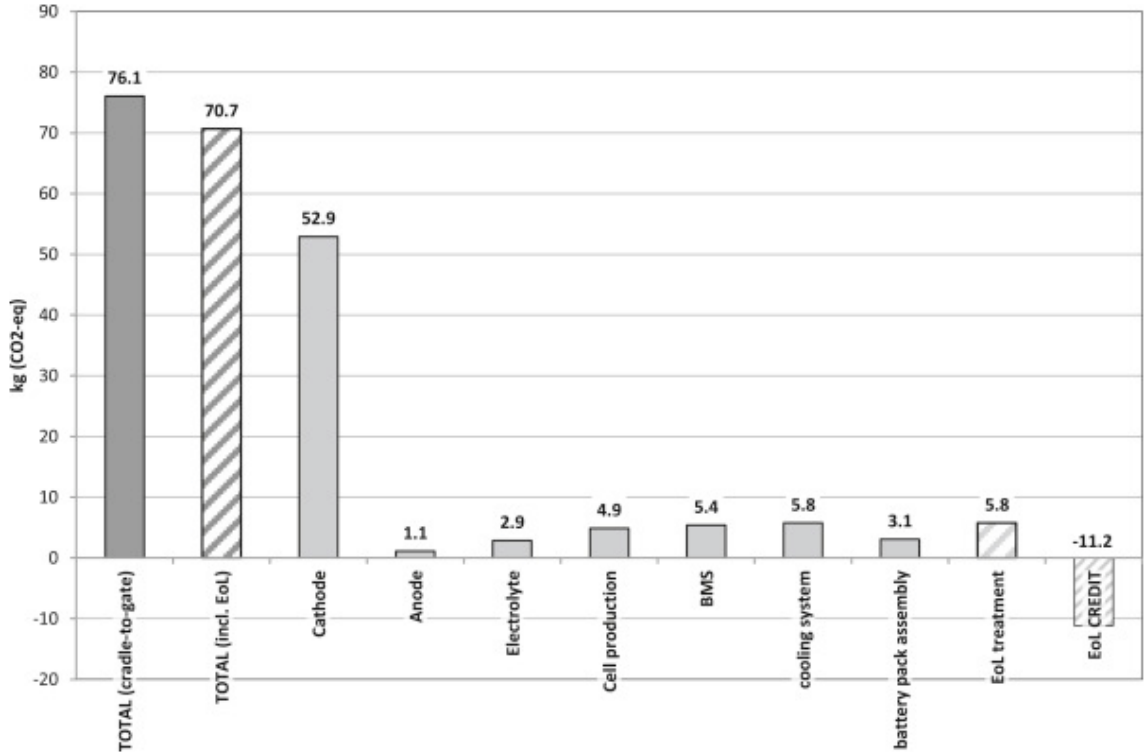

Figure 3 – Cumulative greenhouse gas (GHG) emissions per kWh of battery energy capacity, with and without end-of-life (EoL), and highlighting the individual contributions of the main components (BMS = battery management system).

Here too the cathode stands out as the largest contributor to the overall result, and in fact, its relative share is even higher (~70% of the 'cradle-to-gate' total), due to comparatively large foreground CO_2 emissions resulting from the chemistry of the cathode production process.

When it comes to the EoL, though, the emission credits afforded by the recovery of the valuable metals (all of which are carbon-intensive to produce) lead to a larger relative reduction (-8%) of the total impact in this category.

3.1 Comparison to selected literature

In order to fulfil the second goal of our LCA study, we performed a comparison of our findings to those published in the recent (post-2010) literature for similarly complete 'cradle-to-gate' analyses of commercial battery packs based on the three most common Li-ion chemistries (LMO, NMC and LFP). In particular, seven literature studies were selected as the most representative (Notter et al., 2010; Majeau-Bettez et al., 2011; Dunn et al., 2012¸ U.S. EPA, 2013; Ellingsen et al., 2014; Kim et al., 2016; Hao et al., 2017).

Figure 4 illustrates the results of this comparison, which - due to limitations in some of the literature studies - is confined to the GWP indicator.

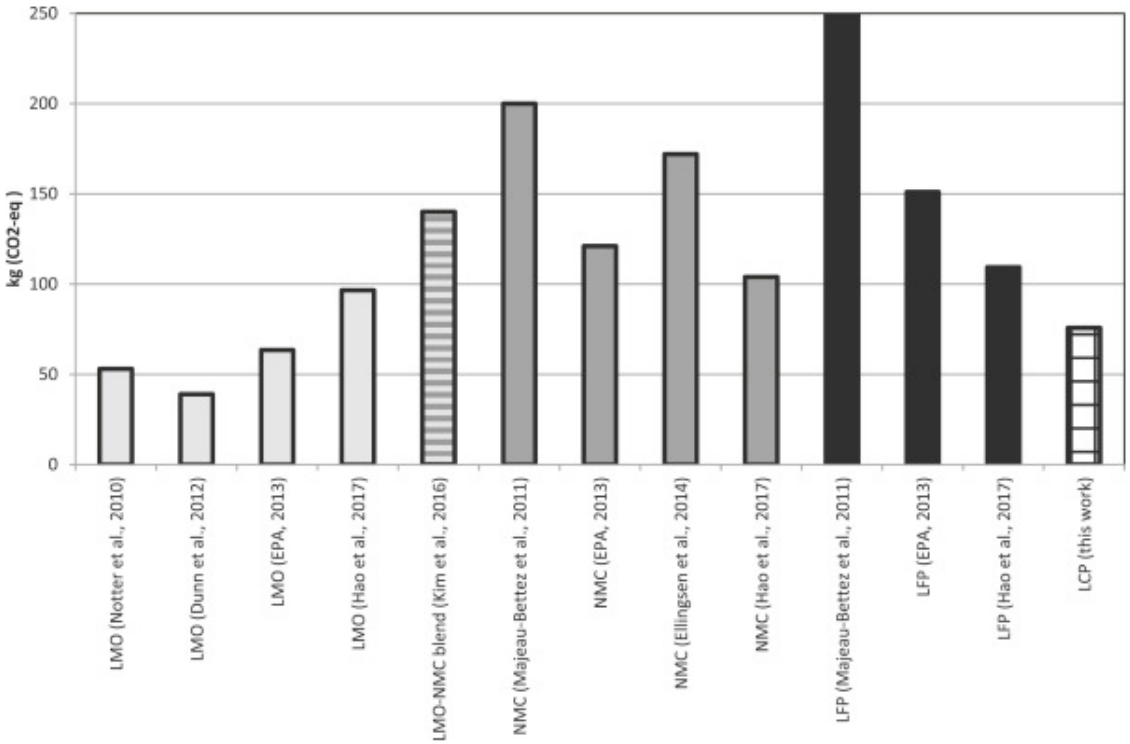

Figure 4 – Comparison of the reported cumulative greenhouse gas (GHG) emissions per kWh of battery energy capacity across selected published studies ('cradle-to-gate' boundary).

The first striking feature of this comparison is the sheer range spanned by the results. On closer inspection, this is due to a number of factors: first and foremost, there is intrinsic variability in the physical size and structure of the analysed battery packs, which leads to wide variations in the assumed relative mass shares of the non-electrochemical components (BMS, cooling system, external packaging, etc.), and therefore on the calculated impacts per kWh of battery energy capacity. Secondly, many of the authors had to rely on "expert estimates", "extrapolations based on R&D values", "top-down estimates", etc., which may have impacted the accuracy of their results. Also, given the demand for high-energy-intensive materials for the production of the cells, the assumed background electricity grid mix(es) and their associated carbon intensities have a potentially large effect on the final results (this is highlighted in Hao et al. (2017), where significantly lower GHG emission values are calculated under the alternative assumption that production could take place in the USA, instead of inChina, to which the values in Figure 4 refer).

Be that as it may, the cathode composition appears to have a distinctive effect on the outcome, as highlighted by the use of different shadings for the bars in the chart. Within each ‘family’ of cathode chemistries, there is some indication of a general downward trend with time (albeit within rather large confidence bands, because of the limitations discussed above, and with the possible exception of LMO batteries).

In light of all of the above, the results for the production of the LCP battery pack analysed here seem to be indicative of a solid performance in terms of total GHG emissions.

4. Conclusions

The LCA of a new type of Li-ion battery pack for EVs based on lithium cobalt phosphate (LCP) chemistry has led to promising results in terms of cumulative energy demand and greenhouse gas emissions. This analysis, and a comparison to previously published results, has also highlighted a number of lingering methodological issues in terms of how to address

and resolve sources of uncertainty linked to battery pack standardization and extrapolation of laboratory and pilot plant inventory data to more meaningful commercial scale operations.

Finally, the performance of a newly developed hydrometallurgical recycling process has also been assessed in a positive light, even though the total reductions in cumulative energy demand and GHG emissions, when also accounting for the credits afforded by EoL material recovery, remain relatively minor.

Acknowledgements

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