



A cradle-to-grave life-cycle-assessment of dry-processed Li-ion batteries for electric vehicles

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Maxwell-type dry processing has emerged as a promising manufacturing technology for high-areal-loading Li-ion battery electrodes, offering a significant advantage by eliminating the use of the toxic and costly NMP solvent. This study developed a cradle-to-grave life-cycle-assessment model to evaluate the environmental impacts of this innovative technology, benchmarking the results against the conventional NMP-based technique for a configured 42 kWh NMC622-graphite battery pack. The findings reveal that the dry-processing method merits a 4.8% lower energy consumption and achieves environmental impacts up to 47.5% lower in 12 out of 13 categories, highlighting its environmental benefits compared with the NMP-based manufacturing processes.

Lifecycle, Electrode, Li-ion battery

1. Introduction

Li-ion batteries (LIBs) are the major energy storage devices for electric vehicles given their high energy and power density, and long cycling life [1-4]. The current manufacturing processes of LIB electrodes are dominated by N-Methyl-2-pyrrolidone (NMP)-based wet processing method, which is expensive, toxic, and energy intensive during the drying and the solvent recovery steps [5, 6]. Additionally, to further improve the energy density of LIBs, one effective way is to increase the electrode active material areal mass loading [7]. Nevertheless, solvent-based thick electrodes usually suffer from binder migration and shrinkage stresses during the heat-drying step. These phenomena not only cause a reduced mechanical strength of electrodes, but also induce increased tortuosity at the electrode surface hindering the electrolyte permeation and undermining the electrochemical performance of LIBs [8, 9].

Recently, researchers have successfully implemented Maxwell-type polytetrafluoroethylene (PTFE)-based dry-processing technique for LIB electrodes with high areal loadings [5, 10-12]. Different from the conventional wet process, the Maxwell-type dry-processing method blends the electrode material components without the addition of solvent. The mixture is then calendered to free-standing electrode sheets and laminated onto current collectors [13]. By employing this dry processing method, all the above-mentioned limitations of the NMP-based wet processing method on thick electrode applications can in theory be addressed [2]. Although the Maxwell-type dry processing method still faces problems such as lower line speed and instability of PTFE at lower potential, the feasibility of this method has been demonstrated in commercialized Tesla 4680 cells [14, 15].

Since the Maxwell-type dry-processing method requires different binder and manufacturing steps, its energy consumptions and environmental impacts need to be investigated and understood comprehensively from a life cycle perspective. Nevertheless, currently there is no life-cycle-assessment (LCA) work ever conducted on dry-processed electrodes for LIBs.

Accordingly, a systematic LCA on this new technology is highly needed to better understand its difference from the conventional solvent-based method. In this work, an LCA model is developed to evaluate the environmental impacts of the dry-processing method for EV LIBs, and the results are compared with the conventional NMP-based manufacturing of LIBs for EVs.

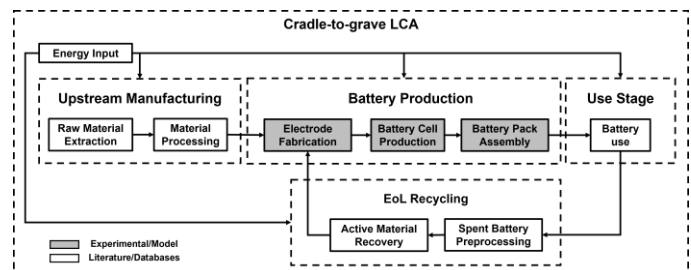


Figure 1. Scope and system boundary for the proposed LCA model

2. Method

A process-based, cradle-to-grave attributional LCA model is developed in this work to evaluate the environmental impacts of the PTFE-based solvent-free electrode manufacturing method [5, 10]. The baseline NMP-based wet processing with PVDF binder is also evaluated for comparison given its dominance in EV battery pack production [2]. The $\text{LiNi}_{0.6}\text{Mn}_{0.2}\text{Co}_{0.2}\text{O}_2$ (NMC622)-graphite EV battery packs for the two processing methods are configured based on a Nissan Leaf battery pack with 192 cells and 40 kWh capacity [16]. Given the energy density increase due to the replacement of $\text{LiNi}_{0.5}\text{Mn}_{0.2}\text{Co}_{0.3}\text{O}_2$ (NMC523) with NMC622 and the enlarged areal loading, the battery pack in this study is configured as 222 kg with 42 kWh of capacity. The scope and system boundary for this cradle-to-grave LCA is shown in Figure 1, including: raw material extraction, material processing, electrode fabrication, battery cell manufacturing, battery pack assembly, battery use, spent battery preprocessing, and active material

recovery. The recovered NMC622 and graphite from the end-of-life (EoL) recycling stage are reused in the electrode fabrication stage.

The functionality is not affected, so the functional unit is defined as a reference flow of 1kg of NMC622-graphite battery pack produced for all material and energy flows. The Life Cycle Inventory (LCI) Analysis is performed using Sphera LCA for Experts (previous GaBi) software version 10.9. The 13 environmental impacts evaluated include Global Warming (GWP), Particulate Matter Formation (PMFP), Fossil Depletion (FDP), Freshwater Ecotoxicity (FETP), Freshwater Eutrophication (FEP), Human Toxicity (HTP), Marine Ecotoxicity (METP), Marine Eutrophication (MEP), Metal Depletion (MDP), Photochemical Oxidant Formation (POFP), Ozone Depletion (ODP), Terrestrial Acidification (TAP), and Terrestrial Ecotoxicity (TETP), and they are characterized using the ReCiPe2016 life cycle impact assessment (LCIA) method.

2.1 Maxwell-type electrode manufacturing for EV batteries

The processes of the dry-processing method are illustrated in Figure 2. As demonstrated in our previous work, both the dry-processed cathode and anode exhibit comparable or enhanced electrochemical performance compared to the NMP-based cathode and anode [5]. Accordingly, we conservatively assume the same active material usage and same performance of the dry-processed and NMP-based battery pack.

For the LCI analysis, the first step of this method is to dry mix active material (NMC622 or graphite), conductive additive (carbon black), and binder (PTFE) in a mass ratio of 92:3:5. For cathode, the components are mixed via a ball mill then processed through a twin-screw extruder. For the anode, mixing is conducted solely on a twin-screw extruder to prevent structural damages to the graphite particles. Afterward, the mixed powder is calendered between hot rolls at 50 °C to form electrode material sheets. The active material areal mass loadings of cathode and anode are set at 30.6 and 17.4 mg cm⁻², respectively [10]. Then a double-layer electrode is produced by laminating two pieces of calendered electrode material sheets onto one piece of current collector through hot rolls at 120 °C. Both the roller speed for calendering and laminating are 1 m min⁻¹. The laminated electrodes are then notched and stacked together for further cell and pack assembly steps. Indeed, the Maxwell-type method can eliminate solvent-related defects such as pinholes, agglomerates, and poor adhesion, though it may also induce other problems such as low edge quality [15, 17]. Due to the limited availability of large-scale production data for the Maxwell-type method, a same scrap rate of 15% is chosen for both methods [17]. The negative to positive (N/P ratio) is assumed at 1.12, and the anodes are 4% larger in area than cathodes for both methods [10, 17]. The driving distance of EVs that use either of the electrode fabrication method is set to 200,000 km at which the capacity of a battery pack would decay to 70% [18]. After the use stage the battery pack is directly recycled using the method described in our previous work [18], whereas the solvent used in the pre-processing of the spent dry-processed electrode has been replaced with 15% Trifluoroacetic Acid (TFA) solution [19]. The recycled graphite and NMC active materials with a collection rate of 91% [16] are reused in the electrode manufacturing steps.

2.2 NMP-based electrode manufacturing for EV batteries

The conventional NMP-based manufacturing processes are illustrated on the left portion of Figure 2. The active material (NMC622/graphite), conductive additive (carbon black), and binder (PVDF) are mixed with NMP solvent in a mass ratio of 92:3:5. In this study, we assumed PVDF the same as PTFE due to

their similar synthesis processes [20, 21]. The cathode and anode solid contents are set at 55% and 45% [17]. The electrode slurries are double-side-coated on the positive and negative current collectors. The coated electrodes are then dried and the NMP vapor from this drying step is recovered and reused in the mixing step. The NMP recovery rate and energy consumptions are 96% and 8.5 kWh kg⁻¹ [22, 23]. The dried cathode and anode are then calendered, notched, and stacked for the following cell and pack assembly processes. In the calendering process, the roller temperature is set at 60 °C [24] and the speed is set at 1 m min⁻¹, the same as for the dry-processing method. The use stage of the EV with NMP-based electrodes is set the same as for the dry-processed electrodes. The EoL recycling method for the spent NMP-based EV batteries is described in our previous work [18]. The NMP solvent usage is adjusted according to this new configuration. Like the dry-processing method, the recycled NMC622 and graphite are reused to produce new battery cells.

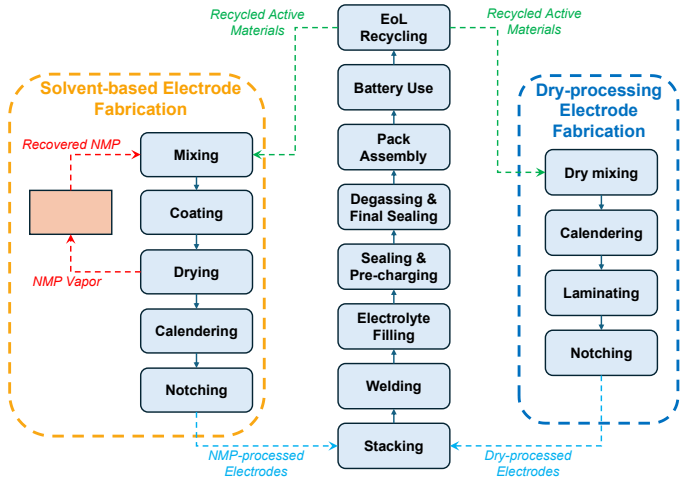


Figure 2. Comparison of EV battery pack manufacturing using solvent-based or dry-processing electrode fabrication methods.

3. Results

The analysis results are presented and discussed in three sections, which include life-cycle-inventory, energy consumption, and environmental impacts of the two manufacturing methods.

3.1 Life-cycle-inventory of the configured EV batteries

Table 1 Mass of components in the configured EV battery pack

Pack Components	NMP-based [kg]	Dry-processed [kg]
NMC622	72.06	72.06
Graphite	42.62	42.62
Carbon Black	3.74	3.74
PVDF	6.23	-
PTFE	-	6.23
Solvent usage (recovered)	142.01 (136.33)	-
Aluminum current Collector	3.17	3.17
Copper current collector	10.97	10.97
Electrolyte	12.64	12.64
Separator	3.34	3.34
Multilayer pouch	0.72	0.72
Terminals	0.72	0.72
Management system	3.01	3.01
Cooling	13.11	13.11
Packaging	49.76	49.76

The mass of each component in the configured wet- and dry-processed EV batteries is listed in Table 1. In the configured

battery, cathode active material NMC622 occupies the highest portion of the total weight, followed by battery packaging and anode active material, graphite. To build the configured EV battery pack, the actual usages of electrode materials including NMC622, graphite, carbon black, binders (PVDF or PTFE), and current collectors are $\sim 17.65\%$ higher due to the 15% scrap rate [17]. When we consider the recycling and reuse of the active materials, with a collection rate of 91% of the spent pack [16], only 19.2 and 11.4 kg extra of NMC622 and graphite are required to build another battery pack. The NMP needed for the solvent-based method is at 142.01 kg for the configured battery pack, whereas 96% of it can be collected and reused, reducing the net NMP usage to 5.68 kg. The weights of current collectors and separators are estimated based on the thickness, area, and density. The electrolyte usage is calculated as 0.3 kg kWh^{-1} of battery pack [25]. Finally, the rest of the components are calculated based on our previous research work [23].

3.2 Energy consumption comparison of the configured EV batteries

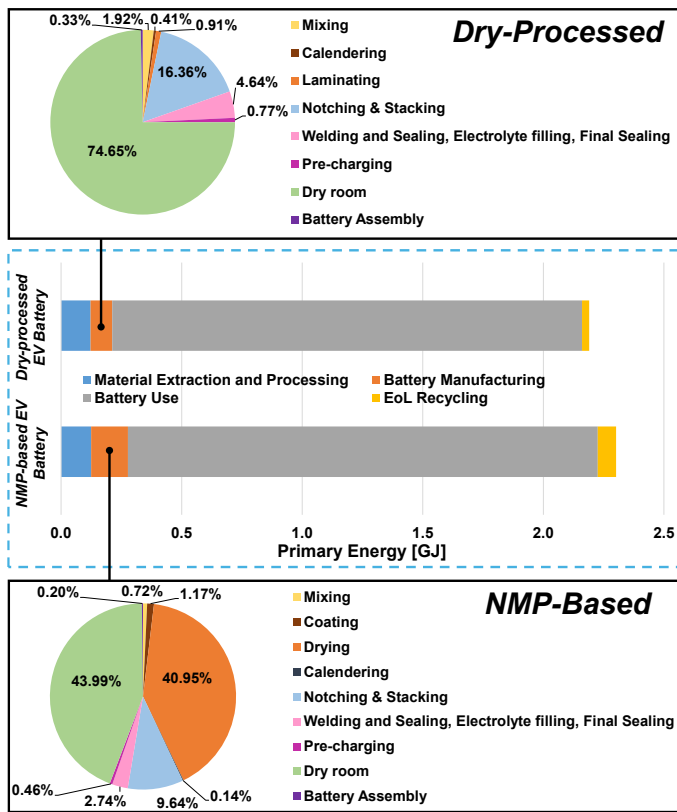


Figure 3. Cradle-to-grave primary energy consumption comparison of the NMP-based and dry-processed EV battery packs

The cradle-to-grave energy consumption is analyzed for the NMP-based and dry-processed EV battery pack and is illustrated in Figure 3, with the contribution of each manufacturing process in the two methods. The total primary energy of the NMP-based and dry-processed EV packs are 2.30 and 2.19 GJ kg^{-1} , respectively. The primary energy consumptions are divided into four stages: Material extraction and processing, Battery manufacturing, Battery use, and EoL recycling. Among all the stages, the battery use stage consumes most of the primary energy at 1.95 GJ kg^{-1} for both the NMP-based and dry-processed battery pack. The material extraction and processing stage consume 124.4 and 122.0 MJ kg^{-1} for NMP-based and dry-processed battery packs, respectively. The difference mainly arises from the production of solvent for the

NMP-based method. In the battery manufacturing stage, the primary energy requirement for a dry-processed battery pack drops to 90.2 from 152.9 MJ kg^{-1} of the NMP-based approach, equivalent to a 41% reduction. Among the NMP-based pack manufacturing processes, the dry room, identified as the top-contributor, consumes 44.0% of the energy, followed by the drying step, which accounts for 41.0% of the energy consumption. In the dry-processed method, the elimination of solvent usage and drying makes dry room the only primary contributor to total energy consumption, accounting for 74.7%. Notably, even though the calendaring and laminating in the dry-processing method use elevated temperatures at 50°C and 120°C , the energy consumptions of these two steps are minor, only contributing to 0.41% and 0.91% of the total manufacturing energy consumption, respectively. In the recycling stage, NMP-based and dry-processed battery packs consume 76.3 and 29.5 MJ kg^{-1} of energy. During the EoL recycling, the main difference between these two methods is from the preprocessing step, specifically from the separation of the electrode materials from the current collectors. Instead of using NMP to dissolve binder, TFA solution was used for the separation in spent dry-processed EV battery pack. TFA has a much lower boiling point than NMP, resulting in much less energy consumption during solvent recovery.

3.3 Environmental impacts of the dry-processed EV battery

The life cycle impacts from the EV battery pack, produced through dry-processed processes, are benchmarked against those of the baseline battery pack produced with NMP-based processes. As illustrated in Figure 4, this comparison spans 13 impact categories for a reference flow of 1 kg battery pack. The dry-processed battery pack has slightly lower impacts in 12 out of the 13 impact categories, and comparable result in HTP (39.656 vs. $39.648 \text{ kg 1,4-DB eq.}$, dry-processed vs. NMP-based). The slightly higher HTP value of the dry-processed method can be attributed to TFA usage during the EoL battery preprocessing. Although the energy consumption during the manufacturing stage of the dry-processed battery pack is lowered by 41%, the advantage is not as significant in the whole life cycle. The primary contributors to all these impact categories are either the material extraction and processing stage or the battery use stage.

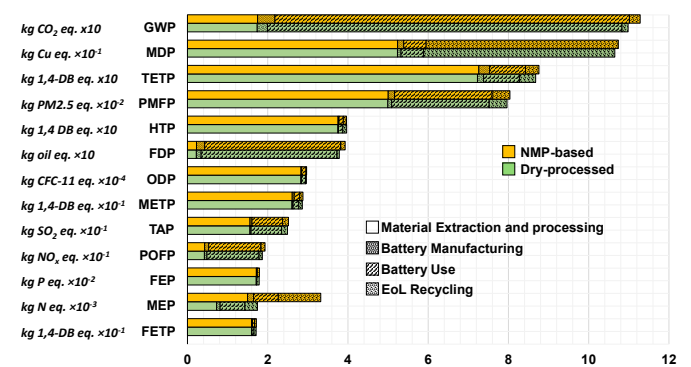


Figure 4. Benchmarking of cradle-to-grave LCA of the NMP-based and dry-processed battery pack

The most significant difference is found for the Marine Eutrophication (MEP) (NMP-based and dry-processed values 3.3×10^{-3} and $1.7 \times 10^{-3} \text{ kg N eq.}$), primarily driven by the NMP solvent usage in the electrode manufacturing and EoL recycling steps of the NMP-based method. The wastewater of NMP production is rich in NMP and monomethylamine, which are degraded to ammonium and nitrate, causing marine

eutrophication [26, 27]. The elimination of NMP solvent in the dry-process method, as a result, reduces the life cycle MEP by 47.6%.

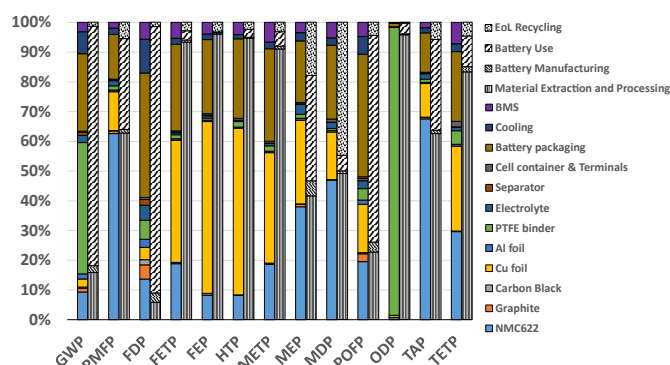


Figure 5. Contribution analysis of the life cycle impacts of the dry-processed battery pack

In Figure 5, the impact contribution of each component during the cradle-to-grave life cycle is shown. It is worth noting the significant size of the green bars in GWP and ODP, which represent PTFE. A similar pattern is observed for PVDF binder in the NMP-based system. The present synthesis paths for TFE and VDF (monomers of PTFE and PVDF) are still highly relying on hydrochlorofluorocarbons (HCFCs) [21, 28], which have significant ODP impact although way lower than the original Chlorofluorocarbons (CFCs). VDF production requires HCFC-142b [28], whereas TFE requires HCFC-22 [21]. Moreover, the production of these HCFCs can also generate hydrofluorocarbon (HFC) by-products (HFC-143a from HCFC-142b production [29] and HFC-23 from HCFC-22 production [30]), with much lower ODP but high GWP, making PVDF or PTFE the top-contributors of GWP among all battery pack components.

Overall, the environmental impacts of the dry-processed EV battery pack from the lab scale are lower than those of the conventional NMP-based counterpart. Other than the significant reduction in MEP (-47.47%) and comparable value (+0.02%) in HTP, the Maxwell-type dry-processing method can shed 0.3% to 3.7% off from the other 11 impact categories compared with the NMP-based method, making it a greener and more sustainable manufacturing option for the high-areal-loading LIB battery pack. Furthermore, it has been demonstrated in literature that a dry-processed $\text{LiNi}_{0.5}\text{Mn}_{1.5}\text{O}_4$ (LNMO) electrode can have superior capacity retention over an NMP-based electrode in full cell cycling test (80% vs. 67% at 300 cycles) [11]. The environmental impacts of the dry-processed battery pack will have even more advantages if its enhanced cyclability is considered.

4. Conclusions

A cradle-to-grave LCA model is developed for a 42.5 kWh NMC622-graphite EV battery pack produced using the Maxwell-type dry processing method. The LCA results are benchmarked against the traditional NMP-based EV battery pack with the same capacity, weight, and lifespan. Results indicate that the dry-processed battery pack has advantages in both energy consumption and environmental impacts. The primary energy of the dry-processed EV battery pack is 2.19 GJ kg^{-1} compared to 2.30 GJ kg^{-1} of the NMP-based battery pack, equivalent to a 4.8% reduction over the whole life cycle of the battery pack. The dry-processed battery pack performs better in 12 of 13 assessed impact categories, suggesting a better environmental sustainability performance. It should be mentioned that the application of Maxwell-type dry-processing for LIB electrodes is

still new. Drawbacks such as lower line speed [5, 17] and poorer performance for the graphite anode still need to be addressed [31]. Nevertheless, its advantages in energy consumption and environmental impacts still makes it a strong alternative to the conventional NMP-based manufacturing technology.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Yu Gu: Methodology, Investigation, Formal analysis, Data Curation, Writing - Review & Editing, Writing - Original Draft, Visualization. **Runming Tao:** Writing - Review & Editing. **Chris Yuan:** Conceptualization, Resources, Supervision, Funding acquisition, Writing - Review & Editing. **Hong C Zhang:** Writing - Review & Editing. **Michael Hauschild:** Writing - Review & Editing.

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