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Abstract: Since environmental benefits and supply chain resilience are commonly assumed for circular economy strategies, this study tests this hypothesis in the context of lithium-ion battery recycling and cell manufacturing. Therefore, the use of recyclates from different cathode active materials and from different recycling routes, namely hydrometallurgy and direct recycling, in a subsequent cell production is modelled with the recyclate quotas prescribed by the amended European Battery Regulation and analysed using life cycle assessment methodology. This study concludes that both, negative and positive environmental impacts can be achieved by the usage of recyclates, depended on the cell technology and the recycling process chosen. Newly constructed lithium iron phosphate (LFP) cells using a share of 11.3% of recyclates, which are obtained from LFP cells by a hydrometallurgical process, achieve a deterioration in the ecology by 7.5% for the global warming potential (GWP) compared to LFP cells without any recyclate share at all. For the same recyclate quota scenario, hydrometallurgical recyclates from lithium nickel manganese cobalt oxide cells (NMC), on the other hand, achieve savings in GWP of up to 1.2%. Recyclates from direct recycling achieve savings in GWP for LPF and NMC of a maximum of 6.3% and 12.3%, by using a recyclate share of 20%. It can be seen that circular economy can raise large savings potentials ecologically, but can also have a contrary effect if not properly applied.

**Keywords:** circular economy; recycling; life cycle assessment; lithium-ion batteries; hydrometallurgy; direct recycling; cell manufacturing; European Union; Battery Regulation

# 1. Introduction

Climate change predicted as early as 1981 by Hansen et al. [1] primarily due to the consistently increasing atmospheric CO<sub>2</sub> concentration is forcing us to decarbonise our actions today [2–4]. The decarbonisation targets for the European Union are laid down in the European Green Deal [5] and call for complete climate neutrality by 2050. While greenhouse gas (GHG) savings have already been achieved in the energy, private households and industry sectors in particular, a stagnation in GHG emissions can be observed in the transportation sector despite increasingly efficient combustion engines [6]. Therefore, the most extensive electrification possible, especially in individual transport, is imperative in order to be able to achieve the European target at all [7–9]. In addition to the urgently necessary GHG reduction across all sectors, the European Union is also heading towards a shortage of critical battery raw materials due to its high exports of high technology goods and the resulting dependence on upstream strategic raw materials [10].

Therefore, circular economy promises to strengthen resilience to supply shortages and geopolitical crises [11]. The British economists Pearce and Kelly [12] introduced in 1990 basic macroeconomic concept of circular economies, in which, after completing their life cycle, resources are not degraded to waste, but are used as long as possible and then returned to the cycle with the help of high-quality recycling processes, which are therefore the key technologies to enable this circularity [13,14]. In addition, it is assumed that



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**Copyright:** © 2022 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). the provision of less greenhouse gas-intensive recyclates can substitute more greenhouse gas-intensive primary raw materials and replace energy-intensive waste treatments [15]. Especially in the context of the decarbonisation of the transportation sector through the market penetration of battery electric vehicles (BEV), expanding the battery life cycle as much as possible [16,17] followed by efficient disassembly [18] and recycling of spent lithium-ion batteries (LIB) is elementary [19]. The critical metals contained on the cathodes of LIBs not only play a strategically crucial role in the European Union (EU) [20], they are also responsible to a large share of the GHG emissions associated with the production of BEVs [21]. At this point, it is important to secure environmentally friendly battery raw materials in the long term and sustainably through effective and efficient recycling.

Due to the recent relevance of recycling LIBs, science and industry increasingly intensify their efforts on further development and improvement of battery recycling processes [22]. But European politics is also reacting in this field with their proposed amended version of the Battery Regulation [23]. In order to legitimately strengthen the recycling of batteries, the European Union sees recycling as the only permissible way of treating spent traction batteries at their end of life (EoL) and also demands minimum proportions of recyclates in new batteries starting from 2030 including a mandatory CO<sub>2</sub> footprint declaration of battery system distributed in the EU [23]. In the map of battery recycling process technologies, a basic distinction can be made between two different routes: direct and indirect battery recycling [24]. While indirect recycling aims to separate the cathode metals deconstructed from the organic components [25,26] of the batteries through metallurgical process steps for new cathode active material synthesis; direct recycling attempts to avoid this segmentation and aims to recover the cathode active materials (CAM) non-destructively and without morphological change [27,28]. Since those CAMs are critical for cathode production, which are produced using solid-state or hydrothermal synthesis, recycling of these materials is highly desirable. This is due to the partly critical precursor materials such as lithium or cobalt salts, but also to the energy required for the synthesis [21,29].

Since its first application for the Coca Cola Company (Atlanta, Georgia, USA) in 1969 [30], life cycle assessment (LCA) has been able to establish itself as a comprehensive scientific quantitative method for the ecological assessment of anthropogenic activities, industrial processing in particular, from a life cycle point of view including: manufacturing phase (begin of life, BoL), use phase and EoL phase [31]. Since the allocation of environmental impacts of the EoL phase of goods plays a decisive role, it can have a decisive influence on the result and must always be coherent with the LCAs goal and scope definition [32]. Hence, two methodological approaches have become commonly used: the avoided-burden and the cut-off allocation method [33]. With the avoided-burden approach, the end-of-life phase is taken into account and attributed to the original product system. Secondary raw materials gained in this way are rewarded with ecological credits. While at the cut-off approach, on the other hand, recycling processes in the end-of-life phase are not attributed to the original product system, but are understood as burdens for the provision of raw materials for the subsequent product system. Here, it makes particularly sense when the production using recyclates is also in the focus of the investigation and thus shows to what extent the use of recyclates can pay off ecologically [33].

While the technological advantages and disadvantages, as well as the ecological performance of the various recycling routes, have already been demonstrated in many studies [34–39], the question of ecological advantage in comparison to primary materials in the context of subsequent fresh battery cell production remains unclear, especially with regard to the amount of recyclates demanded by the EU. For this reason, these ecological effects of recycled materials are to be quantified in this study with the help of life cycle assessment. The cut-off approach is used for this purpose, as the focus of the study is on the ecology of the production of fresh cells using recyclates from the various recycling routes; followed by a comparison with production without additive recyclates.

# 2. Materials and Methods

This LCA is structured according to the standards of ISO 14040 [40] and 14044 [41] and can thus be divided into the four phases: Goal and Scope Definition, Life Cycle Inventory (LCI), Life Cycle Impact Assessment (LCIA) and Interpretation. The goal and scope definition is described here in Section 2.1. The LCI, represented as a detailed description of the technologies considered is presented in Sections 2.2 and 2.3. Since this study is based on data from secondary sources, the data sources are shown in Section 2.4. Results Section represents the LCIA, while the Discussion Section represents the Interpretation.

## 2.1. LCA Goal and Scope Definition

This study presents a consequential LCA [42], which is intended to show what ecological influence can be expected from various types of recycling with subsequential cell manufacturing in the ecosystem of end-of-life lithium-ion batteries by the regulatory provisions at the European level. The aim is to compare the following materials:

- cathode recyclates from industrially established (mechanical pre-treatment in combination with hydrometallurgical conditioning [36,37]) and
- cathode recyclates from recycling technologies in their piloting phase and still under development (direct recycling of cathode active materials [35])
- cathodes from exclusively primary raw material extraction and with the recyclates mentioned before.

The system under investigation is shown in Figure 1 in the form of the system boundary. As the cut-off approach for EoL allocation is used in this study, the recycling of spent batteries is part of the product system of the battery cell manufacturing, and therefore represents the raw material supply for the recyclates used for cell manufacturing. Necessary primary raw materials are also included. Necessary auxiliary and operating materials are also taken into account, as is the subsequent treatment of waste produced at recycling and manufacturing. By-products from the respective recycling processes, such as materials from the module and cell periphery, are not included, nor is their subsequent recycling or utilization treatment. The LCA model is divided into two main parts here: recycling and cell manufacturing. For recycling, the functional unit (FU) is represented by 1 kg of the respective recyclates. For subsequent cell production, the FU is represented by 1 kWh cell capacity and thus represents the reference flow as well. Since the recycling of spent batteries also produces by-products in addition to the cell materials, it is important to describe the procedure for allocating the resulting ecological burdens. In this study, these burdens are allocated at equal shares to recycling outputs (recyclates) as described in Section 2.5. Since the co-products are not included in the system boundary, the environmental impacts are not allocated to them either. This is justified by the fact that the other co-products or co-product materials are not fully recycled, to an extend that they could flow back directly into the production of new goods. Hence, they are considered as waste leaving the system boundary. The recycling processes, as well as the subsequent cell production, are modelled in Germany. The life cycle assessment software GaBi (version 10.6.0.110) by Sphera Solutions, Inc. (Chicago, IL, USA) with the Professional database version 2022.2 was used for modelling. The impact assessment was carried out using the impact-oriented CML2001 [43] method.

The consideration of impact categories in this study is limited to the two global ones of the global warming potential (GWP) and the abiotic resource depletion potential of elements (ADP). These two are important in the context of lithium-ion batteries due to the associated decarbonisation with transport electrification, but at the same time the often-criticised high GHG emissions during the battery production and use of critical raw materials. The acidification potential (AP) and the eutrophication potential (EP) have been chosen as local impact categories, as these categories are also interesting, especially with regard the problems caused by eutrophication in agriculture in parts of Europe [44].



Figure 1. System Boundary of this Life Cycle Assessment.

## 2.2. Recycling Technologies

In recycling metallurgy, i.e., indirect routing, a combination of pyrometallurgical separation of the metals from the organic fraction and subsequent hydrometallurgical processing is typical [45]. Pyrometallurgical pretreatment is considered robust because it can handle a wide range of cell material. However, this is offset by the relatively high inefficiency and high energy requirements [46]. The metals remaining in the alloy mass after pyrometallurgy can thus be subsequently precipitated as sulphates (salts of sulphuric acid) or carbonates (salts of carbonic acid) by dissolving them in acids and alkalis (hydrometallurgy). Further developed indirect processes create a pre-sorting of the target metals through mechanical treatment and thus avoid the pyrometallurgical process step. Thus, after mechanical treatment, the input raw materials can be recovered more efficiently through hydrometallurgical processing. The indirect route achieves end products of very high purity and quality, so that these can be used again as precursors in the production of new battery cells [37,38]. However, due to the complex and complicated process sequence, the recovery rate (recycling efficiency) is not equally high for all the elements involved. Lithium in particular is not possible to recover completely still today.

The mechanical-hydrometallurgical route first crushes the batteries in a shredder under an inert nitrogen atmosphere. Subsequently, the solvents of the electrolyte (dimethyl carbonate DMC and ethylene carbonate EC) can be vaporized at temperatures around 100-140 °C and can be recovered by condensation. Meanwhile, the conductive salt Lithium hexafluorophosphate (LiPF<sub>6</sub>) decomposes and generates organic fluorophosphates, nitric acid (HNO<sub>3</sub>), and hydrofluoric acid (HF) in a gaseous aggregate state, which need to be captured in a gas purification system [47]. Due to the solvents, other substances can also be formed, such as e.g., CO<sub>2</sub>, ethylene (CH<sub>2</sub>CH<sub>2</sub>), and others [48]. Subsequently, the crushed bulk material is separated into the different fractions with the help of sieves, sifters, magnetic and eddy current separation [25]. The so-called black mass, consisting of the electrode coatings, which is separated from the remaining fractions, can then be further processed in the hydrometallurgical treatment.

Hydrometallurgical processing basically comprises three steps: leaching, purification and separation of the target metals. During leaching, the black mass is dissolved in sulphuric acid ( $H_2SO_4$ ) at approx. 40–60 °C [26]. Only graphite remains in solid form and can thus be separated. After leaching, the target metals are purified and separated by solvent extraction, which is used typically for NMC black mass. Cobalt and nickel can be separated and recovered by controlled crystallisation [26]. Lithium is recovered as lithium carbonate ( $Li_2CO_3$ ) in a precipitation process, and can be converted to lithium hydroxide (LiOH) by electrolysis and crystallisation [25]. The data sets of Mohr et al. [36] used in this study differentiate for mechanical pretreatment and hydrometallurgical treatment in the respective cathode chemistries, lithium nickel manganese cobalt oxide (Li-NMC or NMC,  $Li_aNi_xMn_yCo_zO_2$ ) and lithium iron phosphate (LFP). LiOH is recovered directly from cells with NMC cathodes, whereas  $Li_2CO_3$  can be separated in cells with LFP cathodes, which must then be converted to LiOH.

The direct waterjet-based recycling process dismantles the deep-discharged battery modules in a semi-automated process down to electrode level, thus bypassing the crushing and mixing of different fractions such as metals, plastics and organic components. Cell opening takes place in an encapsulated environment, and the exhaust air is subjected to air purification. In this process, part of the solvent from the electrolyte is lost in the air purification. Subsequently, the cathode, consisting of the aluminium collector foil and the electrode coating, is separated into its components in a water jet decoating process. The electrode coating is filtered off, dried and finished [35]. The NMC and LFP recyclates from water-jet-based battery recycling have not undergone any subsequent processing to date. Alternative direct recycling processes use relithiation (e.g., solid-state [49], hydrothermal [50], electrochemical [51] or ionothermal relithiiation [52]) to compensate for the lithium losses during the life cycle and subsequent tempering to regenerate the crystal structure of the cathode active material (CAM).

## 2.3. Cell Manufacturing Technologies

The NMC cathode active material production is divided into three sub-processes: precursor production, solid-state synthesis and post-treatment. In co-precipitation, the three reactants nickel sulphate (NiSO<sub>4</sub>), manganese sulphate (MnSO<sub>4</sub>) and cobal sulphate (CoSO<sub>4</sub>) a66re present in separate solutions. Depending on the desired stoichiometry, the three solutions are combined and subjected to a precipitation with rapid stirring. Subsequently, the heterogeneous NMC mixture must be filtered, washed and dried in order to be able to process it further using the solid-state reaction method. Within the solid-state synthesis, the NMC precursor is mixed with lithium hydroxide (LiOH) and lithium carbonate (Li<sub>2</sub>CO<sub>3</sub>) by means of a dry mixing process. This is followed by the process of calcination via a furnace, which produces the final chemical composition of the NMC. Since calcination sometimes produces larger solid particles, these must be ground and classified in the subsequent step using an air jet mill to produce a powder with a homogeneous size distribution [53].

The cathode material lithium iron phosphate (LiFePO<sub>4</sub>) required for the LFP cell is produced via hydrothermal synthesis. The hydrothermal reaction is carried out in a Parr reactor by mixing iron sulphate salt (FeSO<sub>4</sub>  $\cdot$  7H<sub>2</sub>O) with phosphoric acid (H<sub>3</sub>PO<sub>4</sub>) and lithium hydroxide (LiOH) in water at about 150–200 °C for 5 h. The LiFePO<sub>4</sub> precipitate is collected by suction filtration and then the cathode material is dried in a vacuum oven for several hours [54,55].

For electrode production, first the manufactured NMC active material is mixed with carbon black, a solvent (n-methyl-2-pyrrolidone, NMP) and a binder (polyvinylidene fluoride, PVDF) to form the cathode slurry. The anode electrode production process mixes the graphite with the solvent (deionised water), the binder (carboxymethyl cellulose, CMC) and additives to form an anode slurry. In the second step, the coating, the resulting cathode slurry is applied to the aluminium foil and the anode slurry to the copper foil. The electrodes are then dried in a dryer in which the solvents dissolve at temperatures of 80–160 °C. Once the material is dry, it can be calendered and rolled up again for cutting the coated electrodes to the appropriate sizes for the cell design. Under dry conditions, the cut electrodes are vacuum dried for 12 h before they can be used in the cell assembly. The drying process is the most energy-intensive process in electrode manufacturing [21,53,56].

In the packaging process, the individual sheets (coated on both sides) of electrodes and separator are stacked in a repeating cycle of anode, separator, cathode, separator and so on for prismatic cells. After packaging, the electrolyte is filled into the cell under vacuum with the help of a high-precision dosing needle and sealed afterwards. Last but not least in formation, the manufactured cell is charged and discharged for the first time. The gas produced by the initial charge is removed from the cell via a gas pocket in the degassing process. Ageing is the last step in cell production and is used for quality assurance. During ageing, the cell properties and cell performance are measured by regularly measuring the open circuit voltage (OCV) of the cell over a period of time. Before the cells leave the factory, they are tested in an EoL test stand [21,53,56].

For the energy density of the cells, we calculate 140  $Wh\cdot kg^{-1}$  and 190  $Wh\cdot kg^{-1}$  for LFP and NMC cells, respectively [57].

## 2.4. Data Sources

The life cycle inventory data sets used for the modelling in GaBi of the LIB cell recycling and manufacturing processes described in Sections 2.2 and 2.3 are listed in Table 1 with their data sources. For the modelling of these unit processes, available data sets from the GaBi Professional database version 2022.2 (reporting date: February 2022) have been used. Unit process data sets origin in Germany are therefore used. The same applies to the energy supply processes, such as electrical and thermal energy. If none were available here, European ones were used and if none were available here either, then global ones. LCI is listed in the Supplementary Materials Table S11.

Table 1. Data sources used for LCI.

Unit Process	Description	Source
Advanced Hydrometallurgy	Mechanical Separation, Leaching and Precipitation	[36,37]
Direct Recycling	Waterjet-based Recovery	[35]
NMC Synthesis	Solid-State Synthesis	[21]
LFP Synthesis	Hydrothermal Synthesis	[55]
Cell Manufacturing	Electrode Production, Coating, Drying, Calendering, Packaging and Formation	[58]
Cell Composition	LFP, NMC111 and NMC811, Energy Density	[21,29,37,57]

#### 2.5. Allocation of Burdens, Assumptions, and Scenarios

Because this study pursues the goal of manufacturing battery cells with recycled content, the distribution (allocation of burdens) of the environmental impacts caused by recycling is critical. The allocation of environmental impacts to several products (recyclates) does not follow scientific laws, but represents a subjective distribution key, which has to be well considered [59]. Since the recycling processes considered here are all primarily intended to recover the battery raw materials, the environmental impacts created have only been distributed to these. By-products are therefore not charged with any environmental impacts because they are not fully processed and therefore do not have the necessary quality for direct reuse. When distributing the environmental impacts among the generated battery raw materials, the environmental impacts and the recyclates, as illustrated in Figure 2. Subsequently, an individual footprint for 1kg of the respective recyclate can be calculated by scaling.

Since the waterjet-based direct recycling is still under development, it must be assumed that the recyclates from the waterjet-based recycling can be used for fresh cell production despite the fact that there is no revitalization of the CAM conducted. For the subsequent modelling, scenarios are designed that comply with the amended battery regulation (EU) No 2019/1020 of the European Union [23]. This requires the use of secondary raw materials of lithium, nickel and cobalt in the proportions described in Table 2. In addition, secondary manganese is used to the same extent as secondary cobalt in order to improve comparability with the secondary active materials used from direct recycling. Other battery raw materials, such as graphite or the electrolyte solvents, obtained through hydrometallurgy, are not taken into account in fresh cell production, since they are not required by the upcoming battery regulation. This results in the scenarios for LFP cells



with recyclates from hyrometallurgy (H): *LFP-H-EU1* and *LFP-H-EU2* and with recyclates from direct recycling (**D**): *LFP-D-EU1* and *LFP-D-EU2*.

**Figure 2.** Schematic diagram of allocation of burdens to recyclates and by-products with exemplary numbers.

**Table 2.** Defined scenarios for the production of fresh cells according to the targets of the European Union Battery Regulation [23].

Scenario	Mandatory From	Cobalt Share	Nickel Share	Lithium Share	Manganese Share *
Ref	-	0%	0%	0%	0%
EU1	01.01.2030	12%	4%	4%	4%
EU2	01.01.2035	20%	12%	10%	10%
EU2	01.01.2035	20%	12%	10%	10%

\* There are no mandatory manganese contents for traction batteries in den EU battery regulations but are taken into account for LCA modelling.

Consequently, also for NMC811 cells with: *NMC-H-EU1*, *NMC-H-EU2*, *NMC-D-EU1* and *NMC-D-EU2*.

# 3. Results

## 3.1. LCIA of Battery Recycling

The recycling of NMC and LFP cells results in different recyclates (from hydrometallurgy: Li<sub>2</sub>CO<sub>3</sub> from LFP cells; LiOH, NiSO<sub>4</sub>, MnSO<sub>4</sub> and CoSO<sub>4</sub> from NMC cells and for direct recycling: LFP, NMC111 and NMC811 recyclates) as shown in Figure 3. The allocation of environmental impacts described in Section 2.5 was applied. The functional unit (FU) represented here is described as 1 kg of the individual recyclates. Due to the equal share allocation and subsequent scaling to equal quantity, the environmental impacts are distributed identically in the different impact categories (GWP, AP, EP and ADP). Besides that, it can be seen that the  $Li_2CO_3$  from the hydrometallurgical recycling of LFP cells has the greatest environmental impact. This is due to the lower amount of recyclates generated by hydrometallurgy compared to NMC cells. The precursor recyclates (NiSO<sub>4</sub>, MnSO<sub>4</sub> and NiSO<sub>4</sub>) from NMC111 cells also show the same distribution, corresponding to their approx. equal occurrence. In comparison, LiOH recyclates have about twice the environmental impact the MnSO<sub>4</sub> and NiSO<sub>4</sub> recyclates, as they are also only present in approx. half the mass in the NMC111 cells. For NMC811, the LiOH performs ecologically about the same as for NMC111, but MnSO<sub>4</sub> and CoSO<sub>4</sub> from NMC811 generate significantly higher environmental impacts than from NMC111, due to the significantly reduced amount in the cells. NiSO<sub>4</sub> from NMC811 is accordingly associated with lower environmental impacts than its counterpart from NMC111 cells. In direct recycling, water jetting is the process step

with the greatest environmental impact [35]. For all recyclates from the direct recycling process, the environmental impact values are roughly comparable. The small differences are ultimately due to the different amounts of active material per cell from the cell compositions used (NMC111 [21], NMC811 [37] and LFP [29]). To compare the LCIA results of these recyclates with those of pristine materials, their LCIA is shown in the Supplementary Materials Figure S4.



\*  $CO_2$  = Carbon dioxide;  $SO_2$  = Sulphur dioxide;  $(PO_4)^3$  = Phosphate; Sb = Antimony

**Figure 3.** Life cycle impact assessment of recyclates from battery cells with different cathode active materials (LFP, NMC111 and NMC811) recovered through different battery recycling routes (advanced hydrometallurgy and direct recycling). The functional unit is defined as 1 kg of each of the different recyclates.

# 3.2. LCIA of Cell Manufacturing

The impact assessment for cell manufacturing (Figure 4) using the technologies and data sets mentioned in Section 2 according to the described methodology shows a global warming potential (GWP) of 49.3 kg CO<sub>2</sub>-e·kWh<sup>-1</sup> and 63.2 kg CO<sub>2</sub>-e·kWh<sup>-1</sup> for LFP (*LFP-Ref*) and NMC (*NMC-Ref*) for the reference scenarios. Overall, LFP cells are less greenhouse gas intensive per kWh of cell capacity than NMC cells. For LFP, the admixture of recyclates from the hydrometallurgical recycling route increases the GWP by 1.4% CO<sub>2</sub>-e·kWh<sup>-1</sup> (*LFP-H-EU1*) and 7.5% CO<sub>2</sub>-e·kWh<sup>-1</sup> (*LFP-H-EU2*). The reason for this is primarily the lower yield of recyclates from LFP cells through hydrometallurgical processing and the applied allocation of burdens to recyclates. Recyclates from the direct recycling process, on the other hand, achieve a reduction in global warming potential of -3.6% (*LFP-D-EU1*) and -6.5% CO<sub>2</sub>-e·kWh<sup>-1</sup> (*LFP-D-EU2*). For NMC cells, both EU scenarios through the hydrometallurgical route, approx. -1% CO<sub>2</sub>-e·kWh<sup>-1</sup> (*NMC-H-EU1*) and -1.2% CO<sub>2</sub>-e·kWh<sup>-1</sup> (*NMC-H-EU2*), as well as by direct recycling, approx. -7.3% CO<sub>2</sub>-e·kWh<sup>-1</sup> (*NMC-D-EU2*) can reduce GHG emissions associated with cell manufacturing.



\* CO2 = Carbondioxide; SO2 = Sulphur dioxide; (PO4)3- = Phosphate; Sb =Antimony

**Figure 4.** Life cycle impact assessment of cell production using specific recyclates. The functional unit in each case is 1 kWh of cell capacity. Cells with different amounts of recyclates are shown in each case. The different amounts are defined by the EU scenarios (*EU1* and *EU2*), described in Section 2.5, Table 2. These recyclates origin from different recycling routes: hydrometallurgy or direct recycling (described in Section 2.2). Presented are cells with NMC811 cathode active material and LFP cathode active material.

For the acidification potential (AP), there occur differences in the cell technologies. NMC cells (1.7 kg SO<sub>2</sub>-e·kWh<sup>-1</sup> for *NMC-Ref*) consistently show significantly greater impacts than LFP cells (0.27 kg SO<sub>2</sub>-e·kWh<sup>-1</sup> for *LFP-Ref*). This is due to the manufacturing process of the cells, where the primary raw materials required for the production of the cathodes harbour a significantly greater acidification potential in the case of NMC cathode powder compared to LFP powder, mainly due to the precursor NiSO<sub>4</sub>.

For the eutrophication potential (EP), the picture is reversed with  $0.1 \text{ kg } (\text{PO}_4)^{3-} \text{e-kWh}^{-1}$  (*LFP-Ref*) and  $0.03 \text{ kg } (\text{PO}_4)^{3-} \text{e-kWh}^{-1}$  (*NMC-Ref*). Here, the phosphate emissions into the environment caused during LFP synthesis are the decisive drivers. Overall, it can be seen that the use of hydrometallurgical recyclates in LFP cell production results in an increase in environmental impacts for acidification, eutrophication and abiotic resource depletion potential, as the Li<sub>2</sub>CO<sub>3</sub> recyclate has a greater environmental impact in all impact categories considered here than pristine Li<sub>2</sub>CO<sub>3</sub> from brine for LFP. Only the LFP recyclates from direct recycling can always reduce the environmental impacts (GWP, AP, EP and ADP). For NMC cells, both hydrometallurgical recycling and, to a greater extent, direct recycling can reduce the environmental impacts in these impact categories in the EU scenarios.

For the impact category of abiotic resource depletion potentials (ADP), the same effects are shown for the LFP cell through recyclate from hydrometallurgy (increase of about 10.6% Sb-e), only significantly more pronounced than for GWP, AP and EP. The reason for this is the high resource input required for the wet-chemical treatment, especially through the use of sulphuric acid (H<sub>2</sub>SO<sub>4</sub>) and sodium carbonate (Na<sub>2</sub>CO<sub>3</sub>). Direct recyclates reduce the ADP in scenario *LFP-D-EU2* by about -2% Sb-e. For NMC cells, direct recycling is worthwhile with savings of about -5.2% Sb-e (*NMC-D-EU1*) and about -9% Sb-e (*NMC-D-EU2*). Hydrometallurgy achieves a lower reduction of about -1.7% Sb-e (*NMC-H-EU1*) and -2.1% Sb-e (*NMC-H-EU2*).

# 4. Discussion

# 4.1. Allocation Methods

Since manufacturing processes rarely produce a single product (output), but several, the environmental impacts associated with the manufacturing process must be redistributed to these outputs (allocation). Allocating environmental impacts in a multi-output system is difficult, since there is no strictly scientific solution and is thus associated with a certain arbitrariness [59]. ISO 14044 suggests a system extension to include the downstream recycling processes of the by-products and consequential allocation of environmental impacts among these [41]. Hence, the allocation method described in Section 2.5 does not fully correspond to the proposed procedure according to ISO 14044, because here the environmental impacts are not allocated to by-products. The reason for this is that the focus of this study is on the recovery of active cell material from various recycling routes and not plastics, aluminium or copper. Additional due to the unknown further treatment of these fractions, they are seen as waste and thus no environmental impacts are allocated to them. Subsequently the environmental impacts of these waste fractions when considered as by-products would be identical for both technologies. Consequently, the relative ratios would not change between recyclates and recycling technologies, but only the absolute values. In economic terms: it would only lead to a balance reduction.

In this study, an equal share allocation among outputs (recyclates to be specific), as, followed by scaling is presented, as shown in Section 2.5. This approach has been chosen, because in this way losses due to recycling inefficiencies are taken into account in the footprint of recyclates. By allocating the environmental impacts to a smaller quantity than expected, the environmental footprints for these products increase when scaled to 1 kg of the individual product (recyclate). An example of this is lithium: only 80% of which can be recovered, and accordingly the total environmental impact for lithium is allocated to only 80% of the mass. Thus, the environmental impacts of individual products increase with increasing recycling inefficiency. On the other hand, it can be criticized that goods that occur in a lesser amount than others are penalised, such as cobalt and manganese in NMC811 cells, whose specific environmental impacts increase simply because they occur lesser than nickel, which is even rewarded in this respect. On the other hand, a massproportional allocation bears the danger of a unreasonable allocation, which is exemplary described in the diamond paradox by Kloepffer et al. [59]: Due to the low concentration of diamonds in the rock, diamond mining leads to very large outputs of rock and very small outputs of diamonds. With mass-proportional allocation, almost all environmental impacts would be attributed to the rock and only very few to the diamonds. In addition, when subsequently scaled to the same quantity as functional unit, e.g., 1 kg of the rock or the diamonds, both products would have an identical ecological footprint. The LCIA is performed with mass-proportional allocation, presented as a sensitivity analysis in the Supplementary Materials (Figures S2 and S3).

# 4.2. Key Parameters and Sensitivity

This LCA is based on key parameters, changes to which could affect the results of this study decisively. In order to examine the significance of changes in these key parameters, they have been subjected to a sensitivity analysis. The two most relevant parameters are: the assumed energy density and the missing steps in the process chain of direct waterjet-based recycling to revitalise the CAM.

The gravimetric energy density used for the assessment was increased and decreased by 15% to assess the significance for the overall result of the study, which can be seen in the Supplementary Materials Figure S1. Since the results shift proportionally by varying the energy density within the sensitivity analysis, the findings of this study for reduction or rise of the impact categories through use of recyclates remain unchanged. Nevertheless, the sensitivity analysis shows an inverse ranking between the different cathode technologies in terms of environmental impact. This is illustrated, for example, in the *EU1* hydrometallurgy scenario: Assuming the lower energy density of LFP and the simultaneously high assumed energy density of NMC, NMC becomes ecologically advantageous in the impact categories GWP and ADP. However, the two further considered categories of EP and AP remain in their ecological ranking due to too high absolute value differences.

As described in Section 2.2, the direct waterjet-based recycling process is still in the development phase. Although the CAM can already be separated and recovered on an industrial scale, no revitalisation of the cathode material has been implemented in the process flow yet. In order to estimate the impact of such a processing step for this study, the GWP calculated in this study is compared with that of a direct recycling process with revitalisation by hydrothermal relithiation and subsequent recritsallisation using short annealing. Comparison shown in Table 3.

**Table 3.** Comparison between the waterjet-based recycling approach and direct recycling with hydrothermal relithiation and short annealing.

	Greenhouse Gas Emissions in kg $CO_2$ -e·kg <sup>-1</sup> Cathode Active Material			
Source	NMC111	NMC811	LFP	
This Study	0.59	0.61	0.51	
Gao et al. [60]	1.28	1.34	1.18	
Xu et al. [50]	1.54	1.61	1.42	
Average of literature values *	1.41	1.475	1.3	

Note: \* Values used for the subsequent sensitivity of additional relithiation.

For the following sensitivity analysis, the average values of the direct recycling processes with hydrothermal relithiation presented by Gao et al. [60] and Xu et al. [50] were used, shown in Figure 5. Accordingly, the cell productions of NMC811 and LFP cathodes are modelled with this average value from hydrothermal direct recycling described in Table 3. This shows an increase in the global warming potential for the described scenarios of <1% in all cases. The key findings from Section 3.2 thus remain unchanged for the global warming potential, however, it can be assumed that this is also valid for the other impact categories since GWP should be affected the most due to the high energy demanding short annealing process step during CAM revitalisation.



Global Warming Potential (GWP)

**Figure 5.** Sensitivity analysis with regard to additionally required relithiation and recrystallisation of the cathode material during direct recycling.

## 4.3. Circular Economy Findings

The main findings of this study are that the environmental impacts of battery cells can be reduced by proportionate admixture of recyclates from battery recycling processes, but in some cases even a greater environmental impact is associated with recyclate usage. This is the case for Li<sub>2</sub>CO<sub>3</sub> recovered from LFP cells by hydrometallurgy. As described in Section 3.2, this is due to the yield of the process for LFP cells. Mohr et al. [36] present a review of several studies comparing hydrometallurgical recycling of LFP cells, which confirm this phenomenon for LFP cells, for example in Ciez et al. [61] and Messagie et al. [62] by showing a mathematical positive net impact of LFP recycling (avoided burdens conducted). Another reason for this is the allocation of burdens to the output streams as described in Section 2.5. Since no burdens were allocated to by-products, this study presents here the greatest possible allocation of environmental impacts across the recyclates recovered. Furthermore, from these results we can conclude that hydrometallurgy for LFP cells probably does not describe the ideal process technology and that there is still potential for development here. Especially in view of the renaissance that the LFP cell is experiencing due to optimised system architectures [63,64]. Since hydrometallurgy is still not economically feasible with NMC cathodes [65], hydrometallurgical processing of LFP cells is still much more difficult to operate economically. This could lead to an increase in the price of batteries.

For the NMC cell, on the other hand, hydrometallurgy proves to be advantageous; here, savings can be achieved in the scenarios described across all impact categories considered. These savings seem to be small with approx. -0.8% in GWP for *NMC-H-EU1*, but in view of the low total recyclate input of approx. 4.4% for the metals of the cathode active materials, they can by no means be denied effectiveness. For the *NMC-H-EU2* scenario, the total recyclate input is about 11.3\%, resulting in savings of about -1.1% in GWP. The results from Section 3.2 show noticeable greater savings in the use of recyclates from the direct recycling process. However, it should also be noted that the cathode active material is recovered non-destructive. This means that in order to achieve the 20% recycled content of cobalt, the other metals of the cathode active materials must necessarily consist of recyclates in this proportion as well. This increases the total recyclate content to 12% in scenario *D-EU1* and 20% in *D-EU2*. For this reason, the ecological advantage of the direct recycling compared to hydrometallurgy must be put into perspective.

This study shows that the European Union's recyclate quotas cannot achieve environmental benefits across the board for all technology combinations. Therefore, recycling research should be intensified to address the problems with recovery of Lithium from LFP cells. However, recycling and subsequent use of these recyclates can still make sense. For the reason that recyclates could mitigate possible supply bottlenecks of Lithium [10], so that negative impacts on ecology could also be accepted. This in turn raises the question of whether the level of recyclate quotas is well chosen. Dusseldorp et al. [66] describe that too ambitious recyclate quotas for virgin cells in such a rapidly growing market as that for LIBs can lead to a supply shortage of recyclates. As a result, manufacturers could only meet these quotas through premature recycling and shorter battery life cycles. Wouldn't it perhaps be more appropriate to have mandatory closed-loops for battery recyclates, which could lead in a lesser use of pristine materials and more circular economy efforts?

#### 5. Conclusions

As more and more sectors intensify their efforts on circular economy activities, the scientific investigation and review of the different circularity options is imperative in order to fully exploit the associated potentials. Life cycle assessment (LCA) proves to be suitable for this purpose. However, conducting this LCA also showed that methodological certainty is not fully given. This is important because the European Union also requires the mandatory assessment of the carbon footprint of battery systems. It seems unlikely that cell manufacturers will use higher proportions of recycled material than specified by the regulations. It must be clear that a limited recycled content also only allows a

limited ecological benefit. Here, the results of this study indicate, environmental benefits through circularity of resources in the lithium-ion battery ecosphere can be achieved under specific conditions. However, since ecological disadvantages are possible as well, the beneficial ones must be ensured by making right strategic decisions. This is not only crucial for the producers of goods during product development, but also to political decision-makers when drafting and passing legislation. These regulations are probably the decisive incentives to realise the developmental steps that are still necessary (e.g., more efficient recycling of LFP cells) to implement a resilient circular economy system on a pan-European basis to diminish environmental burdens.

**Supplementary Materials:** The following supporting information can be downloaded at: https:// www.mdpi.com/article/10.3390/su15010103/s1, Figure S1: Sensitivity analysis for different energy densities for the impact categories GWP, EP, Ap and ADP; Figure S2: Mass-proportional allocation related to the functional unit of 1 kg cell material for the impact GWP, EP, AP and ADP; Figure S3: Mass-proportional allocation related to the functional unit of 1 kWh battery capacity for the impact categories GWP, EP, AP and ADP; Figure S4: Environmental impacts of pristine materials used for cell manufacturing presented for comparison reasons; Table S1: Composition of the battery pack; Table S2: NMC111 cell composition; Table S3: NMC111 cathode paste; Table S4: NMC111 anode paste; Table S5: NMC811 cell composition; Table S6: NMC811 cathode paste; Table S7: NMC811 anode paste; Table S8: LFP cell composition; Table S9: LFP cathode paste; Table S10: LFP anode paste; Table S11: Life Cycle Inventories of used unit processes.

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