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Blecua De Pedro, M.; Herrero-Ponce, C.; De Meaza, I.; Martín-Frax, L.; Seguí-Peidro, C.; Boyano, I.; Yañez Díaz, M. (2023). Environmental and economic assessment of a higher energy density and safer operation lithium-ion cell for stationary applications. *Sustainable Materials and Technologies*. 37. <https://doi.org/10.1016/j.susmat.2023.e00704>



The final publication is available at

<https://doi.org/10.1016/j.susmat.2023.e00704>

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Additional Information

Environmental and economic assessment of a higher energy density and safer operation lithium-ion cell for stationary applications

Abstract:

Europe has made significant progress in decarbonizing the planet by increasing the share of renewable energy, with solar, wind, and water being the primary sources of renewable electricity. Energy storage systems, especially batteries, are critical in integrating high shares of renewable energy and ensuring grid stability. Lithium-ion batteries are the preferred choice due to their acceptable cycle life, safe operation at high voltages, and high energy density. However, there is still room for improvement increasing energy density, safety during operation and producing sustainable batteries. This study has identified new materials that can be incorporated into a high-voltage Co-free spinel cathode cell to enhance its electrical performance and ensure safe operation. These materials include a silicon-containing carbonaceous composite as a long-life high-capacity anode active material and a custom-made porous separator based on PVDF, which facilitates the development of a gel-type electrolyte, improving the cell's safety and providing higher cyclability performance. The introduction of the new silicon-containing carbonaceous composite and the PVDF-based membrane is studied in this work from an environmental and economic perspective. An environmental study is performed using the LCA tool to evaluate the sustainability of the Lithium-ion cell production process. An economic study is also performed to calculate the cost of the cells, including raw materials, transport, production process, and externalities costs related to the production process. The Almagrid cell, which has a safer porous separator and an anode made of silicon-containing carbonaceous composite, showed enhanced safety and similar environmental performance compared to the reference cell. However, Almagrid technology will have the potential to improve its sustainability and cost when in-house synthesized components are optimized and scaled up becoming commercially available.

Keywords (min 6):

1. High-voltage Lithium manganese nickel oxide (LNMO) spinel cathode battery
2. Silicon-containing carbonaceous composite anode
3. PVDF-based separator
4. Gel Polymer Electrolytes
5. Life Cycle Assessment
6. Life Cycle Costing

Abbreviations:

Battery Energy Storage Systems (BESS)

Capital Expenditure (CAPEX)

Critical Raw Material (CRM)

Energy Storage Systems (ESS)

Environmental Impacts (EIs)

External Cost Factors (ECFs)

External Environmental (EE)

External Environmental Cost (C_{EE})

Fine particulate matter formation (PMF)

Fossil resource scarcity (FF)

Freshwater eutrophication (FE)

Gel Polymer Electrolytes (GPEs)

Global Warming (GW)

Greenhouse Gases (GHG)

Human carcinogenic toxicity (HTc)

Human non-carcinogenic toxicity (HTnc)

Ionizing radiation (IR)

Lithium Nickel Manganese Oxide (LNMO)

Labour Cost (C_L)

Land use (LO)

Life Cycle Assessment (LCA)

Life Cycle Impact Assessment (LCIA)

Life Cycle Inventory Analysis (LCI)

Lithium-Ion Batteries (LIBs)

Lithium Iron Phosphate (LFP)

Marine eutrophication (ME)

Mineral resource scarcity (SO)

Nickel Cobalt Aluminum Oxide (NCA)

Nickel Manganese Cobalt Oxide (NMC)

Operational Expenditure (OPEX)

Ozone formation, Human health (HOF)

Ozone formation, Terrestrial ecosystems (EOF)

Poly (Vinylidene Fluoride-Co-Hexafluoropropylene) (PVDF-HFP)

Poly(Methyl Methacrylate) (PMMA)

Polyacrylonitrile (PAN)

Polyethylene (PE)

Polypropylene (PP)

Polyvinylidene Fluoride (PVDF)

Production Costs (C_P)

Raw Materials Cost (C_{RM})

Stratospheric Ozone Depletion (OD)

Supplementary Information (SI)

Terrestrial acidification (TA)

Waste Cost (C_W)

Water consumption (WC)

1. Introduction

Europe is one of the most recognized regions leading the decarbonization of the planet by introducing the use of renewable energies. In fact, according to EUROSTAT sources (1), the share of renewable energy more than doubled between 2004 and 2020 being wind and water the responsible for providing more renewable electricity. In addition, solar is the fastest-growing energy source, over one-fifth of energy was used for heating and cooling from renewable sources, and 10.2% of renewable energy was used in transport activities in 2020. However, different crises have been and are currently affecting the European energy system. Firstly, the COVID-19 pandemic induced a rise in the natural gas cost because of the sudden economic recovery after the inactivity period during the pandemic (2). In this way, the energy supply sector saw a partial switch to more carbon-intensive energy fuels, while the strong growth in renewable energy observed in recent years lost pace in 2021 (3). More recently, because of the 2022 Russian invasion of Ukraine, the European Commission has deployed what has been called the REPowerEU plan action to step up the green transition away from Russian gas by accelerating the deployment of renewables in an attempt to increase the renewable energy's shares and stop the dependence on Russian natural gas. However, the high penetration of variable and intermittent sources such as photovoltaic and wind power presents a number of challenges that must be considered to successfully make the transition from a conventional electricity system based on fossil fuels to a low-carbon electricity system. On the one hand, it must be ensured that the intermittent nature of these sources does not affect the stability and quality of the power grid supply. On the other hand, displacing manageable conventional power plants from the grid will reduce the inertia of the power system and the capacity to supply auxiliary grid services, such as primary and secondary backup, by traditional means. In this context, energy storage systems (ESS), and most specifically battery energy storage systems (BESS), can play a vital role in meeting these challenges and favoring the grid integration of high shares of renewable energies. The relevance of storage has already been observed in the rise of lithium-ion batteries (LIBs) for on-grid applications, being the base technology for most of the current projects under demonstration, providing grid support, frequency regulation, peak shaving, leading to higher reliability, stability, and lower cost to the power grid.

Among BESS, LIBs have proved to be the customer choice because of their acceptable cycle life, safe operation at high voltage, and high energy density (4,5). However, several research and development actions can also improve other aspects of LIBs employed for stationary applications. Among these improvements are reaching higher energy densities using high-voltage cathodes, silicon as an additive in the graphite negative electrode, and the use of high-safety electrolyte and separators. In addition, the production of sustainable batteries has been located on the front line of the technology development, motivated by the Regulation (EU) 2023/1542 of the European Parliament and of the Council of 12 July 2023 concerning batteries and waste batteries, amending Directive 2008/98/EC and Regulation (EU) 2019/1020 and repealing Directive 2006/66/EC. This regulation includes the declaration of the battery carbon footprint which is directly proportional to non-sustainable resources and production practices. Within the broad category of LIBs for utility-scale storage, nickel manganese cobalt oxide (NMC) cathode chemistry has roughly 50% of the market, with lithium iron phosphate (LFP) and nickel cobalt aluminum oxide (NCA) chemistries gaining quickly (7). However, conventional cobalt (Co)-containing cathodes suffer from comparably low abundance, high costs, and supply chain uncertainties associated with this metal. Recycling is unlikely to provide significant short-term supply, hence fostering the search for Co-free alternatives. One of the latest development efforts (8) that aims to improve the cell voltage, and consequently the energy density, is the use of new Co-free cathode alternatives such as the spinel lithium nickel manganese oxide (LNMO) that can increase the cell operating voltage up to 4.7 V vs Li/Li⁺ compared with 3.7 V for NMC cathodes and shows very fast lithium insertion and extraction (9). Therefore, this high-voltage cathode system is an environmentally friendly and a low-cost alternative for safe, high-energy density, and high-power applications (10).

With regards to the anode, graphite is still the active material dominating the market despite of its theoretical Li-storage capacity (372 mAh/g) (11). Silicon has emerged as a solution due to its high theoretical capacity of 4,200 mAh/g (full Li alloying) and low electrochemical potential vs Li/Li⁺ (0.4 V). This increases the chances of developing a practical Si-based anode for LIBs, whose inherent drawback is its nearly 400 % volume change during lithiation (12). The mechanical properties of graphitic or amorphous carbon make it able to buffer the volume expansion of Si, avoiding damage to the cell, while the high conductivity of C can efficiently complement the high lithiation capacity of Si. Although Si-graphite electrodes have been extensively reported owing to their relatively superior electrochemical performance and acceptable cycle life, their wide application has not been realized yet in commercial batteries (13,14). However, graphite is considered a critical raw material and its high level of exploitation due to current and future demand could finally lead to the depletion of natural graphite resources. Moreover, graphite manufacturing is characterized by an energy-intensive production process (15,16). In this context, other carbonaceous materials with no supply issues could be used as graphite substitutes. Carbon materials coming from abundant carbon-rich wastes, such as biomass, are a promising type of materials for their co-utilization with silicon in LIB anodes. Biomass exact composition depends mainly on biomass species, but it is also influenced by other factors like geographical location or seasonal changes (17). The use of biomass can reduce the carbon footprint of the BESS. This strategy is then aligned with a circular economy perspective and the urgent need of developing EES that not only possess an outstanding performance but have also lower costs and increased sustainability (18,19). Several research works have been conducted on the development of these types of materials and their use in LIBs. Different biomass

precursors have been investigated for this purpose including bamboo chopsticks (18), wood chips (20), orange peel (21), avocado seeds (22), wheat bran (23) or corn cobs (19).

Besides the electrodes and cell active components, the separator is also a key component for batteries. Although it is not directly involved in any cell reaction, its structure and properties are decisive in battery performance including cycle life, safety, energy or power density. Separators must be chemically, electrochemically, thermally, and mechanically stable to battery operation conditions and other battery materials. Moreover, wettability by the liquid electrolyte and porosity are properties of utmost importance to ensure electrolyte absorption and retention so that the batteries possess adequate ionic conductivity. Additionally, separators are electronic insulators too so as to prevent electrical short circuit (24–26).

Microporous membrane separators are usually made of polyolefins such as polyethylene (PE) and polypropylene (PP) as well as their multilayer combinations PE/PP or PP/PE/PP. Although these materials have essential properties for their use as separators, their use could be restricted for high battery performance due to their poor thermal stability, low wettability, and poor electrolyte retention. In order to meet requirements of advanced battery generations, other polymeric materials are being used for membrane synthesis, being the most reported ones polyvinylidene fluoride (PVDF) and its copolymer poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP), polyacrylonitrile (PAN) and poly(methyl methacrylate) (PMMA). Among them, PVDF stands out mainly due to its great wettability because of its good affinity to liquid electrolyte solutions. It also possesses other properties such as physicochemical, electrochemical, and mechanical stability in lithium-ion batteries (24,27). Their high wettability and good retention of liquid electrolyte allow the development of PVDF-based gel polymer electrolytes (GPEs), where the liquid electrolyte is immobilized in the polymeric matrix reducing thus the risk of leakage and consequently improving battery safety. GPEs act both as separator and electrolyte and combine advantages of solid and liquid electrolytes. This combination results in high ionic conductivity and good interfacial properties from the liquid phase and good mechanical properties from the solid polymer matrix (28,29). It is therefore a promising solution for the development of a new generation of batteries.

New battery technologies are expected to cause lower environmental impacts compared to current technologies, thus promoting environmentally sustainable development. Nonetheless, the remarkable growing demand for LIBs and different issues related to their whole life-cycle such as battery degradation mechanisms, energy consumption, greenhouse gases (GHG) emissions, or raw materials depletion require in-depth sustainability assessments in order to evaluate possible environmental implications of batteries. Moreover, these issues could also be responsible for health consequences. In this sense, Life Cycle Assessment (LCA) is a powerful tool that provides a holistic approach to determine and overcome these impacts (30). LCA is the compilation and evaluation of the inputs (materials and energy) and outputs (products and emissions) at each step of the battery life considering raw materials acquisition, production, use phase and end-of-life stage including recycling and/or disposal. It supports the identification of hotspots of impacts and the comparison of different options thus assisting decision-making and enhancing the environmental efficiency of the battery manufacturing (31). However, despite its great potential, there is no consensus in the field of LCA on how to analyze the environmental impact of batteries and how the results should be reported. As a consequence, a wide variety of methods are used making

comparisons between different studies difficult. It also limits the ability of LCA to provide reliable feedback for decision making (30,32,33). This problem has been pointed out by several authors and, although it has not yet been addressed, different studies have been carried out for unifying LCA approaches (34). Efforts must be made in this direction in order to make use of the full potential that LCA tool can provide. Even though several studies on LIBs have been conducted, there is a significant lack of detailed LCA studies which consider all the life stages of a battery. Many studies are focused on the impacts of the LIB production, but the use phase and the end of life are not sufficiently addressed. Regarding applications, most studies have focused on the automotive and there are only few studies when it comes to stationary applications (33,35).

According to the ROADMAP ON STATIONARY APPLICATIONS FOR BATTERIES prepared by “Batteries Europe” the cost-competitiveness of the BESS is a key challenge to be addressed to ensure its usability and further development (36). In this sense, one of the targets to address is a cost reduction of 50% for the upcoming batteries, being the current cost 141 €/kWh (37). In this way, the economic analysis of the new battery becomes a key driver for the future development of the technology. Many researchers have performed investigations in this area (38,39). A review from Rahman *et al.* (38), where 91 articles were analyzed, concluded that an increase in the discharge duration reduced the Levelized Cost of Energy (LCOE). The LCOE indicates the selling price that implies a total return of the cost due to the production and maintenance of the BESS (38)). This cost includes Capital Expenditure (CAPEX), Operational Expenditure (OPEX), and more costs such as the balance of the plant, Operation & Maintenance, Replacement, Disposal and Recycling and External Environmental (Zakeri and Syri, 2015; (40)). However, the CAPEX is normally the highest and most significant parameter as can be seen in table 17 from reference (41).

In this study, the following materials have been chosen as new components for the battery cell in order to improve the electrical performance as well as ensure safe operation.

- LNMO: high-voltage and Co-free oxide with stable 3D spinel structure providing higher energy density and durability.
- Silicon containing carbonaceous composite as sustainable high-capacity anode active material and alternative to Si-graphite mixtures.
- Separator: tailor-made PVDF-based porous separator with excellent wettability providing higher cell performance in terms of cycle-life. Its good retention of liquid electrolyte allows the development of gel-type electrolyte improving thus the cell safety.

According to the above-mentioned reasons, this study compares two types of lithium-ion cells from an economic and sustainability perspective. A reference cell made of an LNMO cathode, silicon-graphite anode, and a commercial separator, and an Almagrid cell made of an LNMO cathode, silicon-containing carbonaceous composite, and a safer porous separator based on PVDF. The combination of these components has been tested at lab scale (coin cells) showing promising results. Nevertheless, it has not been upscaled to industrial cell format yet (pouch cell). Due to the important role of sustainability in the upcoming BESS, an environmental study using the LCA tool following a cradle-to-gate approach is performed before up-scaling the manufacturing process. In addition to the LCA, an economic study is also performed, as the battery production cost is one of the most critical parameters, if not the most, prior to

the up-scaling process of a novel technology. The economic evaluation is performed by the calculation of the cost of the cells, including the raw materials, transport, production process, and externalities cost related to the production process. The results of the study will serve as justification to upscale the production process of these types of cells that will increase the high energy density, sustainability, and safety of LIBs for stationary applications, and therefore allow the increase of renewable energy shares in the grid.

2. Materials and methods:

2.1. Cell study cases

Table 1 shows the composition of the Reference and Almagrid cells. As was explained in the introduction, Almagrid cell components have been tested at lab scale (coin cells) showing promising results which have been extrapolated to a 40Ah pouch cell format. In this way, the cell potential and its integration within the industrial cell format can be proved. As can be seen in the table, the difference between both cells is related to the composition of the anode and the separator. In the case of the Almagrid cell, the anode material is based on a silicon-containing carbonaceous composite whilst the reference cell contains a silicon (10 wt%)/graphite blend. In the case of the separator, the selected commercial one for the reference cell is a Celgard 2500 monolayer polypropylene grade. In the case of the Almagrid cell, a novel porous separator synthesized by ITE based on PVDF material is included. More info about the raw materials and their location and providers can be found in **Table S. 1** from the Supplementary information (SI).

Table 1. Reference and Almagrid cells components. Source: CIDETEC cells' composition.

Component	Subcomponent	Reference cell	Almagrid cell	Unit
Anode	Graphite	0.52		kg/kg anode
	Activated carbon		0.12	kg/kg anode
	Silicon	0.07	0.20	kg/kg anode
	Super C45 carbon	0.04		kg/kg anode
	Carboxymethyl cellulose (CMC)	0.04	0.04	kg/kg anode
	Styrene butadiene rubber (SBR)	0.04	0.04	kg/kg anode
	H ₂ O (ink solvent)	1.26	1.95	L/kg anode
	Copper current collector	0.48	0.79	kg/kg anode
Cathode	LiNi _{0.5} Mn _{1.5} O ₄ (LNMO)		0.97	kg/kg cathode
	Super C65 carbon		0.05	kg/kg cathode
	PVDF		0.05	kg/kg cathode
	NMP ink solvent		1.43	L/kg cathode
	Aluminum current collector		0.08	kg/kg cathode
Cell	Anode	147.70	96.99	g/cell
	Cathode	387.00	403.56	g/cell
	Celgard 2500 Monolayer PP	18.85		g/cell
	PVDF porous membrane		82.15	g/cell

Carbonate and Ethyl methyl carbonate (EC:EMC; 1:1 in volume), 1 M lithium hexafluorophosphate (LiPF ₆)	183.46	183.38	g/cell
Aluminum laminated foil	17.14	17.14	g/cell
Terminal tabs	5.50	5.50	g/cell
Total	764.68	791.85	g/cell

More data regarding the design of the cells can be found below:

- Nominal capacity: 40Ah (charge at 0.2C, 25±3°C / discharge at 0.2C, 25±3°C)
- Nominal voltage: 4.4 – 4.5V
- Voltage during operation: 4.8 – 3.5V
- Maximum charge: 40A at 1C
- Maximum discharge: 120A at 3C
- Internal resistance: <3mΩ at AC @1kHz
- Energy density: 235Wh/kg
- Cycles (80% capacity, State of Health): 1000 cycles at 0.5C/0.5C; 80% Depth of Discharge (@25±3°C)
- Dimensions: Width: 146mm; Length: 255mm; Thickness: 8.2mm
- Charge/discharge efficiency (coulombic; CE): >99.5%

Even though both types of cells have the same electrical properties, the main advantage of the Almagrid cell is related to the increased energy density and safety. Laboratory trials in full coin cells containing a PVDF membrane showed an improvement of 10% in the capacity and 43% of increase in cycle life at C/2 and 25°C compared to commercial polyolefin separator. In the case of the 50 wt.% Si-based composite, it was tested at half coin cell and displayed an outstanding capacity of around 1,884 mAh g⁻¹ Si at C/3 rate. However, these results have not been published yet.

2.2. Production process

In general terms, the production process was composed by the following steps:

- **Anode and cathode manufacturing:** Mixing (Slurry preparation in the planetary mixer), Coating (Slurry casting onto CC foil), Drying (Convection oven coupled to coating line), Calendaring (Roll-pressing of the coated CC), Slitting (cutting of the electrodes).
- **Cell assembly:** Stacking/winding of the cell core (Electrodes and separator stack), Tab welding by Ultrasounds, Packaging (Thermal sealing pouch), Cell Vacuum drying in a Vacuum oven, electrolyte filling and sealing (Vacuum chamber + syringe injection + thermal sealing).
- **Cell finishing:** Formation (Electrical charge/discharge), Degassing and sealing (Vacuum chamber + puncture + thermal seal), and Aging/Grading (Electrical test).

More details about the production process of the pouch cells can be found in (42).

2.3. LCA methodology

LCA is a powerful tool to determine the environmental impacts of LNMO-based lithium-ion cells and to establish a comparative study between both cells developed. In addition, opportunities can be identified to improve the environmental performance of the products in the different stages of their life cycle.

LCA has been performed according to ISO standards, specifically ISO 14040 and ISO 14044. In accordance with ISO 14040, Life Cycle is defined as the consecutive and interrelated stages of a product system, i.e., from the acquisition of raw materials or their generation from natural resources to final disposal.

It mainly consists of four stages: (1) Goal and scope definition, (2) Life Cycle Inventory Analysis (LCI), (3) Life Cycle Impact Assessment (LCIA) and (4) Interpretation of obtained results.

– Goal and scope

In the first step, the aim of the study is defined, and main methodological choices are made, such as **functional unit** definition or the identification of the system boundaries.

Functional unit defined for this analysis is a 40Ah Li-ion cell. This kind of cell consists of six main elements: anode, cathode, electrolyte, separator, cell container and tabs. Two types of cells are considered as stated in section 2.1.

For carrying out this analysis Cradle to Gate scope is selected. It covers every step involved in cell development, from raw materials acquisition to cell production including also materials transportation. In this case, product distribution and end-of-life stages are not considered.

– Life Cycle Inventory

LCI stage involves the identification and quantification of inputs and outputs of the previously defined system throughout its life cycle. Inputs and outputs considered are raw materials, energy, products, and co-products as well as waste and emissions to air, water or soil. LCI is mainly based on experimental data for cells production, and it is also complemented with information obtained from literature and databases such as Ecoinvent.

In this case, LCI is structured in 3 stages: raw materials acquisition, materials transportation, and cell production. Electrodes included in the cells are developed during the project and the rest of the components considered are commercial, except for the Almagrid cell separator which is self-produced too. For electrode production, the steps considered are those listed in the previous section.

Cell composition is shown in **Table 1**. Differences observed among the cell composition values are mainly due to anodes capacity as well as separator density.

– Life Cycle Impact Assessment

In this phase, identification and evaluation of the amount and significance of the potential environmental impacts derived from LCI previously compiled are performed. Inputs and outputs identified are classified and associated to different impact categories and indicators. Some examples of impact categories are climate change, acidification, or resource depletion. These categories usually cover three areas of

protection: human health, natural resources, and ecosystem quality. A wide variety of methods could be used to allow this evaluation.

LCIA, therefore, involves the selection and characterization of the most relevant impact categories and the characterisation model selection. For the study of both lithium-ion cells, characterization was performed using the ReCiPe method for midpoint indicators from the hierarchical perspective.

SimaPro software and Ecoinvent database have been used to compile the data and to calculate the impacts of this LCA study.

The main objective of the method that has been chosen in the study is to transform the long list of LCI results analysed through Ecoinvent into a limited number of indicator scores. The scores of these indicators express the relative severity in an environmental impact category. ReCiPe comprises 18 impact categories at the midpoint level (problem oriented), although in this study ecotoxicity categories have been excluded since final values are associated with a large uncertainty (41). Selected impact categories are shown in **Table 2**.

Table 2. Impact categories selected from the ReCiPe method.

Impact category	Unit
Global warming	kg CO ₂ eq
Stratospheric ozone depletion	kg CFC11 eq
Ionizing radiation	kBq Co-60 eq
Ozone formation, Human health	kg NO _x eq
Fine particulate matter formation	kg PM _{2.5} eq
Ozone formation, Terrestrial ecosystems	kg NO _x eq
Terrestrial acidification	kg SO ₂ eq
Freshwater eutrophication	kg P eq
Marine eutrophication	kg N eq
Human carcinogenic toxicity	kg 1,4-DCB
Human non-carcinogenic toxicity	kg 1,4-DCB
Land use	m ² a crop eq
Mineral resource scarcity	kg Cu eq
Fossil resource scarcity	kg oil eq
Water consumption	m ³

– Interpretation

In this phase, the results of the previous phases (LCI and LCIA) are evaluated and interpreted according to the stated goal and scope initially defined. In addition to impacts quantification, conclusions related to raw materials extraction optimization, energy saving processes, minimizing pollution and waste generated could be drawn.

2.4. Economic assessment methodology

In this article, the economic assessment pursues to make a comparison between both cells under study. This study covers the fabrication cost as well as the cost related to the externalities due to the fabrication of the cell. Considerations made for the fabrication process are shown in **Table S. 10 from SI**.

As both cells feature the same design apart from the raw materials, the above parameters are valid for both designs. The costs of the cells are calculated per year and after that, the cost is standardized per kWh using the annual cells' production.

The collected costs regarding the production process and the externalities cost are the following:

- Raw materials cost (C_{RM}): this cost represents the expenditure done by the purchase of raw materials involved in the production process. The main raw materials are the ones that comprised the anode and cathode, as well as the electrolyte, separator, current collectors, and packaging of the cells. This cost also includes the transportation cost from the supplier to the plant located in Donostia, Spain. The prices as well as the origin of these materials are shown in **Table S. 2 from SI**.
- Production costs (C_P): this cost represents the electricity, natural gas or another types of energy used for carrying out the manufacturing processes. In this case, just electricity is used to produce the cells. The cost of electricity is set as 0.08 €/kWh according to some Spanish industrial plants.
- Labour cost (C_L): cost of the personnel hours spent in the fabrication of the cells. The personnel cost is 9.69€/h (43). 6 operators are working per day in the plant during 2 shifts of 8 hours.
- Waste cost (C_W): incremental expense directly attributed to the disposal of production scrap. The defect rate of the production process is 5% for both cells. The waste cost for the scrap coming from the anode, cathode, and cell casing leftovers is 41.22 €/kg, considering this scrap as Waste Electrical and Electronic Equipment (WEEEs), as well as its transport from the plant to the waste management plant. Separator's leftovers are considered within the waste category "Contaminated wipes and absorbents". The cost of their disposal is 153.75€/kg considering their transport in big bags. Electrolyte leftovers are not considered, as it is considered that this residue will be evaporated.
- External environmental cost (C_{EE}): This cost refers to the costs linked to the environmental impacts (EIs) obtained from the LCA. These impacts were converted to economic values using External Cost Factors (ECFs), provided by literature (44). For the present analysis, those values were updated to December 2022 prices by applying the annual inflation rate in Spain according to reference (45) from 2015 to 2021 and reference (46) for 2022. The updated ECFs to 2022 prices are shown in **Table 3**.

Table 3. Updated ECF to monetarism the EI.

Environmental Indicator (EI)	Unit	Environmental price as external cost (12/2022) (€/Unit EI)	Reference year	Reference
Climate change	kg CO2 eq	0.07	2015	(47)
Ozone layer depletion	kg CFC-11 eq	34.75	2015	(47)
Human carcinogenic toxicity	kg 1,4-DB eq	0.11	2021	(48)

Human non-carcinogenic toxicity	kg 1,4-DB eq	0.11	2021	(48)
Ionising radiation	kBq Co-60 eq	0.05	2021	(48)
Terrestrial acidification	kg SO ₂ eq	5.68	2015	(47)
Freshwater eutrophication	kg P eq	2.13	2015	(47)
Marine eutrophication	kg N eq	3.56	2015	(47)
Urban land occupation	m ² a	0.10	2015	(47)
Particulate matter formation	kg PM _{2.5} eq	65.02	2017	(49)
Water scarcity	m ³ water eq	0.09	2017	(49)
Mineral resource scarcity	kg Cu eq	0.19	2018	(44)
Ozone formation, Human health	kg NO _x eq	Data not available.		
Ozone formation, Terrestrial ecosystems	kg NO _x eq	Data not available.		
Fossil resource scarcity	kg oil eq	0.05	2019	(50)

The C_{EE} of each EI ($C_{EE, EI}$) was then calculated as the product between EI result (EI unit/kWh of provided electricity) and ECF (€/EI unit). Finally, the total C_{EE} corresponds to the sum of the single $C_{EE, EI}$.

Finally, the CAPEX was calculated following the below equation:

$$CAPEX \left(\frac{\text{€}}{\text{year}} \right) = C_{RM} + C_P + C_L + C_D + C_{EE}$$

$$CAPEX \left(\frac{\text{€}}{\text{cell}} \right) = \frac{CAPEX}{\text{Annual production}}$$

Economic inputs related to both cells under study were provided directly by the suppliers of the materials, or from the literature. More information can be found in the Supplementary Information. No comparison with commercial cells is described in this article as the manufacturing process for the cells under study is done on a pilot plant scale instead of on an industrial scale.

3. Results and discussion:

3.1. Environmental analysis

Following the methodology described in the previous section, LCA of both cells is performed. Moreover, in depth analysis of silicon/graphite blend and silicon-containing carbonaceous composite anodes is presented so as to better analyse environmental implications of novel anodes development.

3.1.1. Anodes comparison

Inputs considered for inventory compilation are raw materials employed for the electrodes production as well as transport needed for delivering these materials to the electrodes production site (San Sebastián) and electricity required for the anode electrode production process. On the other hand, outputs taken into account are the manufactured anode, the emissions, and the process residues. In this case, emissions are represented by solvent evaporated, i.e. water, and the solid residue is 15% of the total anode produced.

As for the anode composition, as it is shown in **Table 1**, the main difference between both cells is the active material. In the Almagrid cell the silicon/graphite blend has been substituted by silicon-containing carbonaceous composite. Due to the different nature and properties of these materials, the rest of the anode composition has been readjusted too (**Figure 1**).

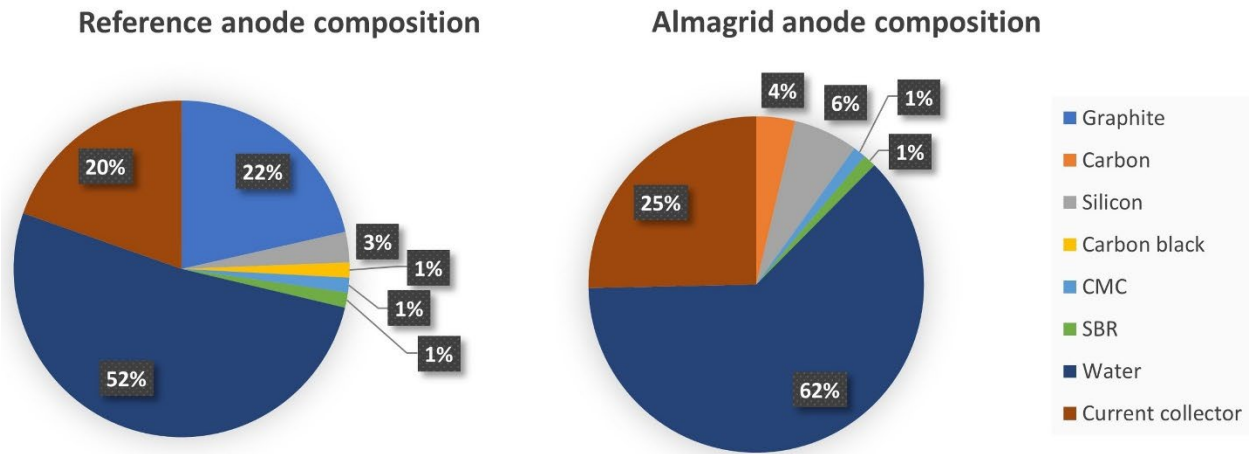


Figure 1. Percentual distribution of the composition of analysed anodes. Source: CIDETEC anodes' composition.

Once the LCI is defined, the LCIA is performed. First of all, LCIA results attained by using midpoint methodology are reported for the complete anode production process. The analysis is mainly focused on global warming (GW) indicator with the aim of identifying the most relevant contributors and looking for alternatives to overcome environmental hotspots.

Reference anode total impact in terms of GW indicator is 37 kg CO₂ equivalent while Almagrid anode impact is 53.9 kg CO₂ equivalent. The difference obtained is remarkable, nonetheless Almagrid anode capacity is higher and thus impact at cell level will be offset given the smaller amount of anode needed.

Figure 2 and **Table 4** show anodes' impact breakdown by raw materials, transport, electricity, and waste. In both cases, reference and Almagrid anode, the trend observed is the same being electricity employed in the production process the main contributor to total impact. This comparison demonstrates that regardless of the anode composition the electricity required for the assembly process is the greatest contributor to the GHG emissions. Electricity is highly dependent on the country's electricity mix considered, and it accounts for approximately 70% of both anodes. Anodes production thus entails high energy demand.

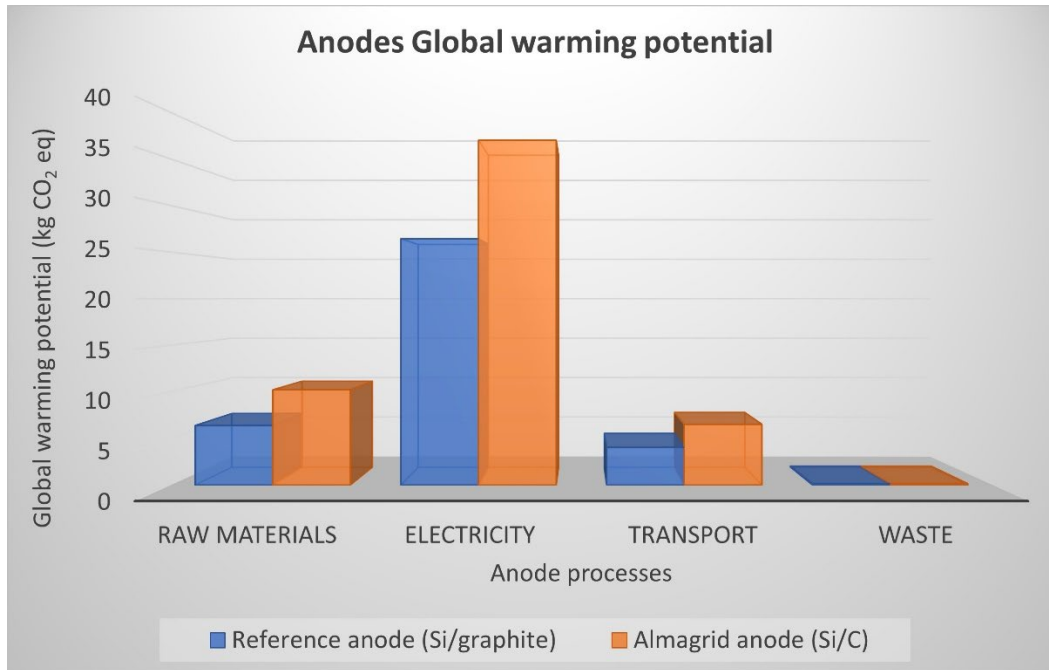


Figure 2. Anodes global warming potential comparative between reference and Almagrid anode. Source: LCA (Simapro software and Ecoinvent database).

Table 4. Global warming impact percentual distribution (%). Source: LCA (Simapro software and Ecoinvent database).

Stage	Reference anode (%)	Almagrid anode (%)
Raw materials	17.3	19.0
Transport	10.9	12.1
Electricity	71.4	68.6
Waste	0.4	0.3

Transport and raw materials production have also considerable contribution to global warming. In the case of raw materials, copper current collector has the highest impact being 75% and 78% for the reference and the Almagrid anode respectively, although its composition share is 20% and 25%. Its impact is the second main anode contribution after electricity representing around 13-15% of the overall impact of the anodes. Copper impact mainly relies on copper mining since it is a highly energy-intensive process.

Although this study is focused on GW indicator other selected indicators have been analysed too. Results for these indicators are presented in **Figure 3**.

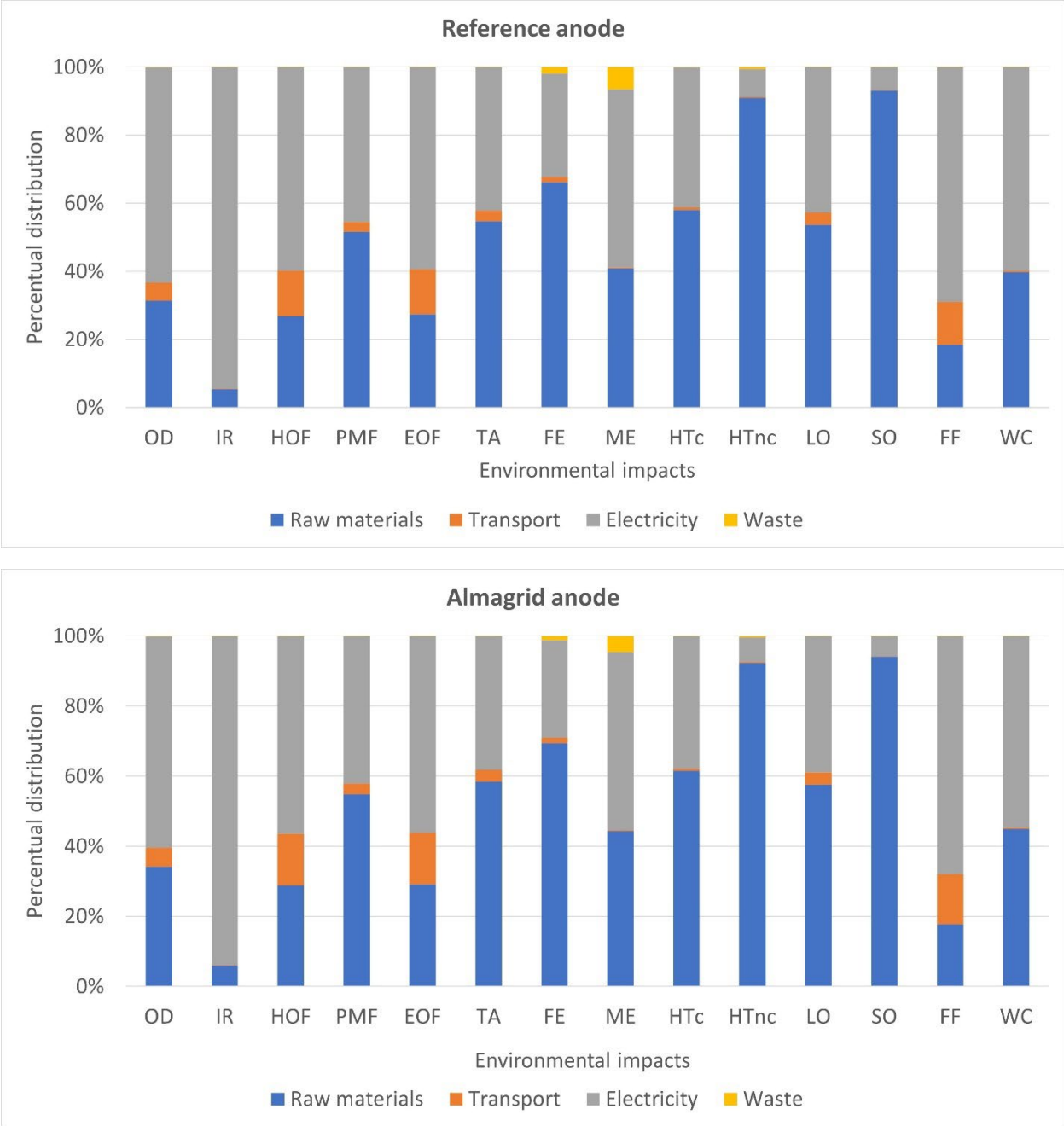


Figure 3. Environmental impacts breakdown for Reference and Almagrid anodes. Source: LCA (Simapro software and Ecoinvent database).

As shown in **Figure 3**, tendencies observed for Reference and Almagrid anodes in terms of environmental impacts are similar. For most indicators, the highest contribution is mainly assigned to electricity or raw materials processing. Especially noteworthy is the contribution of electricity in the case of ionizing radiation, and raw materials for non-carcinogenic toxicity and mineral resource scarcity with contributions higher than 90% for both anodes.

Regarding electricity, it should be noted that the energy mix considered in this study includes coal-based energy, the fact that would explain the contribution of around 70% of electricity to fossil resource scarcity impact. In the case of raw materials, its high impact mostly corresponds to copper current collector. As it was stated previously, mining and processing of copper are highly intensive processes.

It is also worth noting that transport contribution is always less than 15%. Even reduced contribution is found for the waste category, with values lower than 0,5% except for freshwater and marine eutrophication where values around 1-2% and 5-7% respectively are reached.

Analysis of the anodes using endpoint methodology can be seen in the SI (Figure S. 1).

3.1.2. Cells comparison

Regarding cell evaluation, first of all, LCI is collected taking into account every input and output considered for studying LCA of cells production. On the one hand, process inputs are cell components, their transport to cell production site, and electricity consumed for manufacturing process. As it has been previously stated main differences between both cells rely on the anode and the separator. Anodes and cathode have been developed by Cidetec while Almagrid separator has been produced by ITE. The rest of components used for cell implementation are commercial. On the other hand, outputs are the product, namely the cell, and generated waste considering in this case 5% of total cell weight. **Figure 4** shows the percentual distribution of the composition of both cells under study.

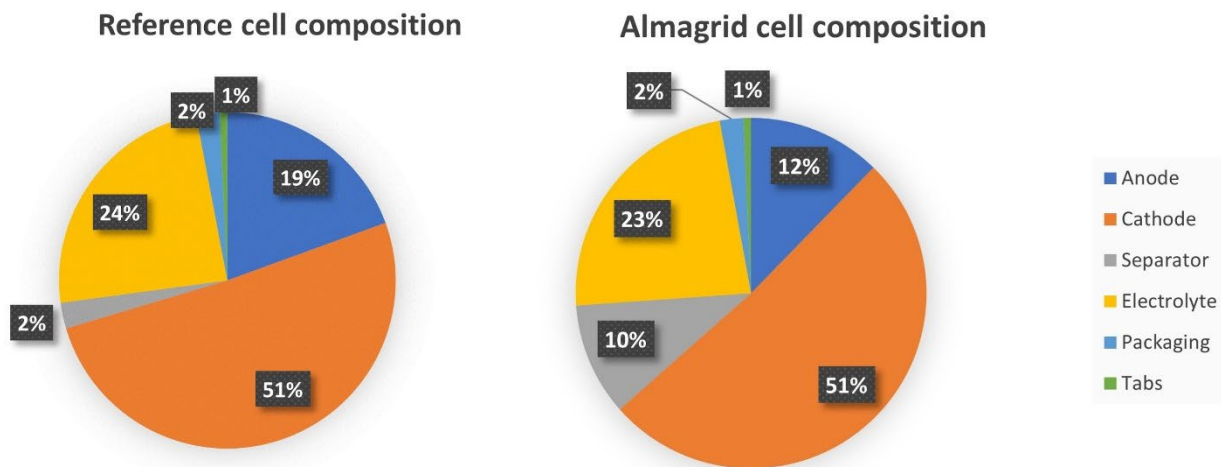


Figure 4. Percentual distribution of the composition of analysed cells. Source: CIDETEC cells' composition.

For cell production environmental assessment, the same analysis as for the anodes has been conducted. GW indicator analysis by using midpoint methodology is performed. Reference cell overall impact is 49.5 kg CO₂ equivalent whereas for Almagrid cell the value obtained is 51.7 kg CO₂ equivalent. In this case, differences between both cells are smaller than for the anodes. As it was previously introduced, Almagrid anode capacity is higher and consequently lower anode quantity is required for cell integration.

Figure 5 illustrates impact breakdown in terms of cell components, transport, electricity and waste and **Table 5** shows the percentual distribution. Cathode, electricity, and anode are the main contributors to total impact accounting for about 60, 25 and 10% respectively. Trends observed are similar in both cases. The most remarkable impact is related to the cathode which has a weight share of 51% of the total cell weight for both cells. The cathode production also entails high energy demand having electricity the biggest contribution (64%) to GW indicator followed by NMP solvent (12%), LNMO-based active material production (11%), and transport of raw materials to manufacturing site (9%).

Concerning the cell comparison, it should also be pointed out that Almagrid separator has a higher impact assigned than the reference one due to higher mass of this separator required for cell implementation because of its superior thickness. Additionally, Almagrid separator has been developed at laboratory scale and its production still needs to be optimized and scaled up, whereas reference separator is already a commercial product. Nonetheless, Almagrid separator is a promising component which has demonstrated superior performance at laboratory scale. Higher performance of this separator should be proved during the use phase, not considered in this LCA. It is expected that taking into account these considerations overall impact of Almagrid cell would substantially decrease.

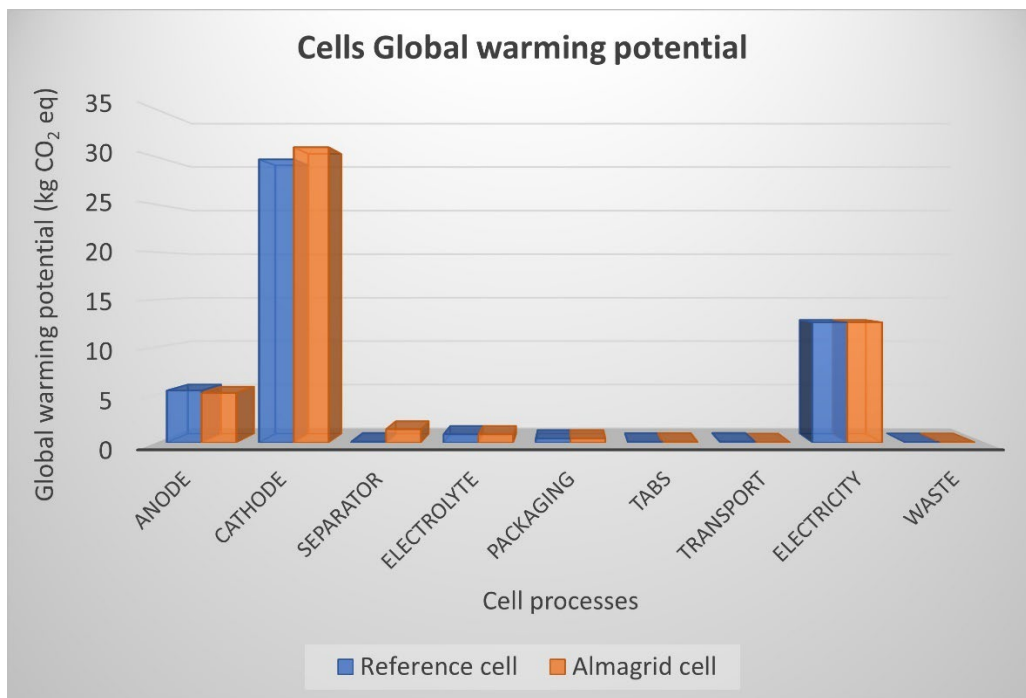


Figure 5. Cell global warming potential comparative between reference and Almagrid cells. Source: LCA (Simapro software and Ecoinvent database).

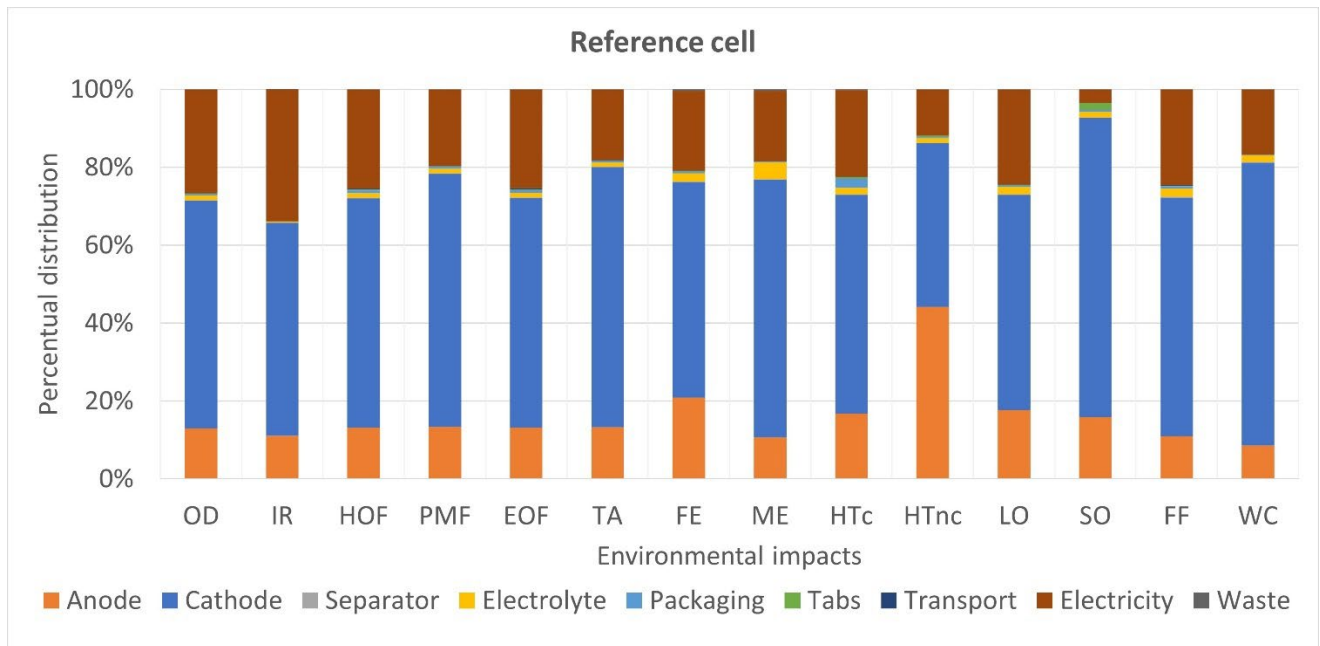
Table 5. Global warming cells impact percentual distribution (%). Source: LCA (Simapro software and Ecoinvent database).

Component	Reference cell (%)	Almagrid cell (%)
Anode	11.1	10.1
Cathode	60.1	60.0

Separator	0.2	2.7
Electrolyte	1.8	1.7
Packaging	0.9	0.8
Tabs	0.2	0.2
Transport	0.2	0.04
Electricity	25.5	24.4
Waste	0.1	0.1

In cells study, extended impacts analysis has also been carried out as shown in **Figure 6**. Trends for both cells in terms of impacts breakdown are the same, although the main differences arise from separator as pointed out in the case of GW indicator. Especially noticeable are the cases of Ozone formation (terrestrial ecosystems and human health) and fossil resource scarcity with Almagrid separator contributions of approximately 4-7%.

The highest contribution to every impact category is assigned to the electrodes, with the main contributors being the current collector and LNMO in the case of anodes and cathodes, respectively. Electricity also represents significant impact, especially for Stratospheric Ozone Depletion and Ionizing Radiation where contributions higher than 20% are reached.



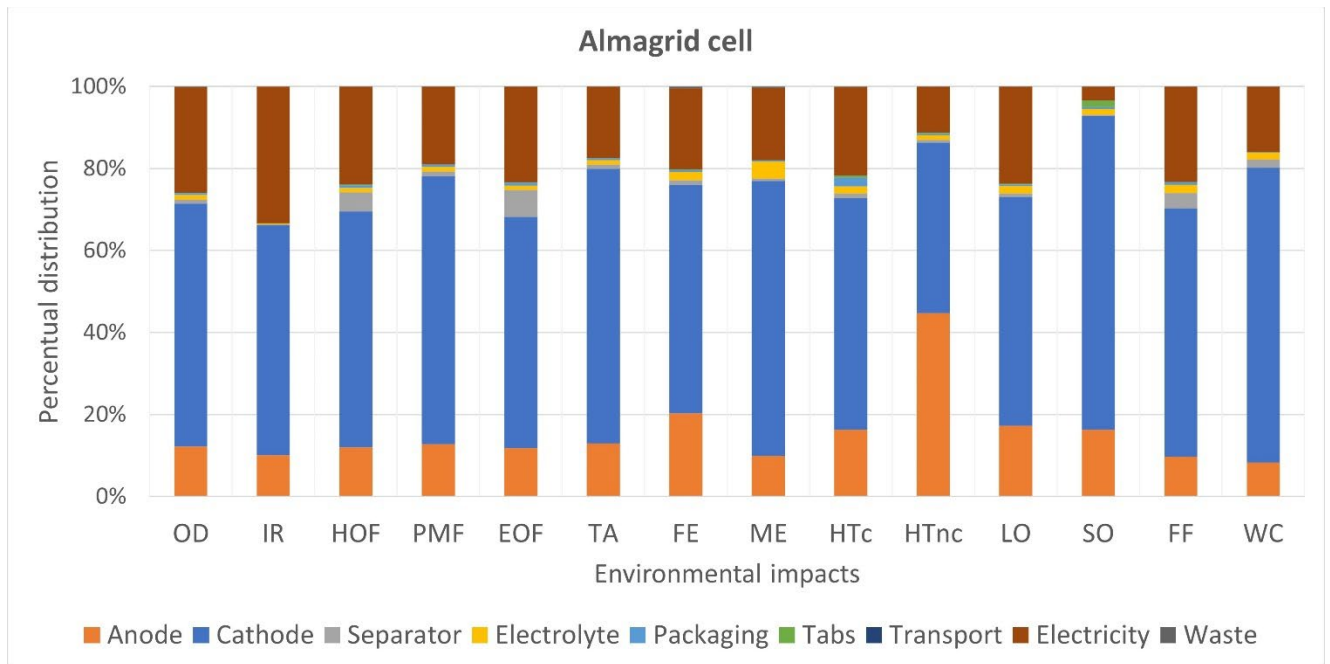


Figure 6. Environmental impacts breakdown for Reference and Almagrid cells. Source: LCA (Simapro software and Ecoinvent database).

3.2. Economic analysis

3.2.1. Production process

The cells under study are produced in CIDETEC pilot plant facilities. This plant operates 16h per day. The production processes are described in **Table S. 9 from the SI. Table 6** shows the daily production amounts. Production of 1,000g of the anode is established for both types of cells, and then the cathode production is adjusted based on the cell requirements. One reference cell comprises 147.70g of the anode and 387.00g of the cathode. One Almagrid cell comprises 96.98g of the anode and 403.56g of the cathode. Due to the fact that less amount of the anode is needed for the Almagrid cell production, more cells are produced at the end of the year, i.e., a 52% production increase is seen when Almagrid cells are produced.

Table 6. Production data for reference and Almagrid cells. Source: CIDETEC pilot plant.

Parameter	Reference cell	Almagrid cell
1-A - Anode manufacturing (kg/day)	1.00	1.00
1-B - Cathode manufacturing (kg/day)	2.62	4.16
2 - Cell assembly (cell/day)	6.77	10.31
3 - Cell finishing (cell/day)	6.77	10.31
Cell production (cells/year)	2,234.26	3,402.48
Cell capacity (Ah)	40.00	40.00
Cell voltage (V)	4.50	4.50
Cell energy (kWh)	180.00	180.00
Cell production (MWh/year)	402.17	612.45

3.2.2. Raw materials cost

Table S. 11 and Table S. 12 from the SI show the raw material cost breakdown for the Reference and Almagrid cells respectively. **Table 7** shows the cost related to the raw materials used for anode, cathode, and cell production. As can be seen in the table, the anode from the Reference cell is more economic than the one from the Almagrid cell. This is mainly due to the higher silicon concentration found in the Almagrid cell, 0.195 kg of Si per kg of anode, versus 0.07 kg of Si per kg of anode in the case of the Reference cell. Furthermore, silicon is the most expensive material, having a cost of 1,870 €/kg. The second most expensive anode component is the activated carbon used in the Almagrid cell, which costs 138.94 €/kg, in comparison with the Graphite used in the Reference cell whose cost is 49 €/kg. However, the amount of activated carbon in the Almagrid cell is 0.117 kg per kg of anode, while the Reference cell employs 0.52 kg of graphite per kg of anode. In the case of the cathode, the cost of this component is the same for both cells, as the same formulation is used for both cells. However, in terms of manufacturing, a higher cost is accounted for the Almagrid cell due to the higher amount of cathode material found in this cell. **Table 7** shows a higher raw material cell cost for Almagrid cell. This difference does not come from the electrolyte, the aluminum foil, and the tabs costs, as the same type and number of materials are used for both cells. The main differences come from the anode costs, and as well as from the separator cost. The Reference cell employs a Celgard 2500 Monolayer PP separator (340.16 €/kg), whereas the Almagrid cell employs an innovative and more expensive PVDF Porous membrane (552.71 €/kg). In addition, more amount of separator is needed in the case of the Almagrid cell (82.15 g/cell) in comparison with the Reference one (18.85 g/cell). The PVDF membrane used in the Almagrid cell is still under development, meaning that the amount of material needed will be lower in the future because of the use of thinner separators, as well as the industrialization of the production process of these membranes.

Table 7. Raw material costs of the different cell components. Source: LCC.

Component	Raw materials cell cost (€/cell)	
	Reference cell	Almagrid cell
Anode	26.76	40.09
Cathode	58.20	60.69
Cell	174.94	229.73

Figure 7 shows the breakdown of the raw material costs in anode, and cells for the Reference and Almagrid cases. The shares of the different components in the case of the cathode are the same for the Reference and the Almagrid case. The main anode components are the silicon and the graphite or activated carbon (see cell composition in **Table 1**). According to the explanations from the former paragraph, it can be seen in the anode graphics (**Figure 7 upper part**) how the silicon cost is higher for the Almagrid cell (88% vs. 74%). In conclusion, the Almagrid anode cost mainly comes from the higher silicon concentration. In the case of the cell costs, the graphic shows a higher share for the separator in the case of the Almagrid cell (20% vs. 4% for the Reference cell) due to the higher amount of separator used in this cell, and its more expensive price. The anode share is higher in the Almagrid cell due to the higher amount of silicon employed in this cell. The shares of the rest of the components are different from one cell to another due to the final cost of the cells. However, it can be appreciated how in the case of Almagrid cell the use of silicon and of PVDF porous membrane increase the cell cost at the point as the electrolyte share decays to 33%, 10% less in comparison with the Reference cell (electrolyte share: 44%).

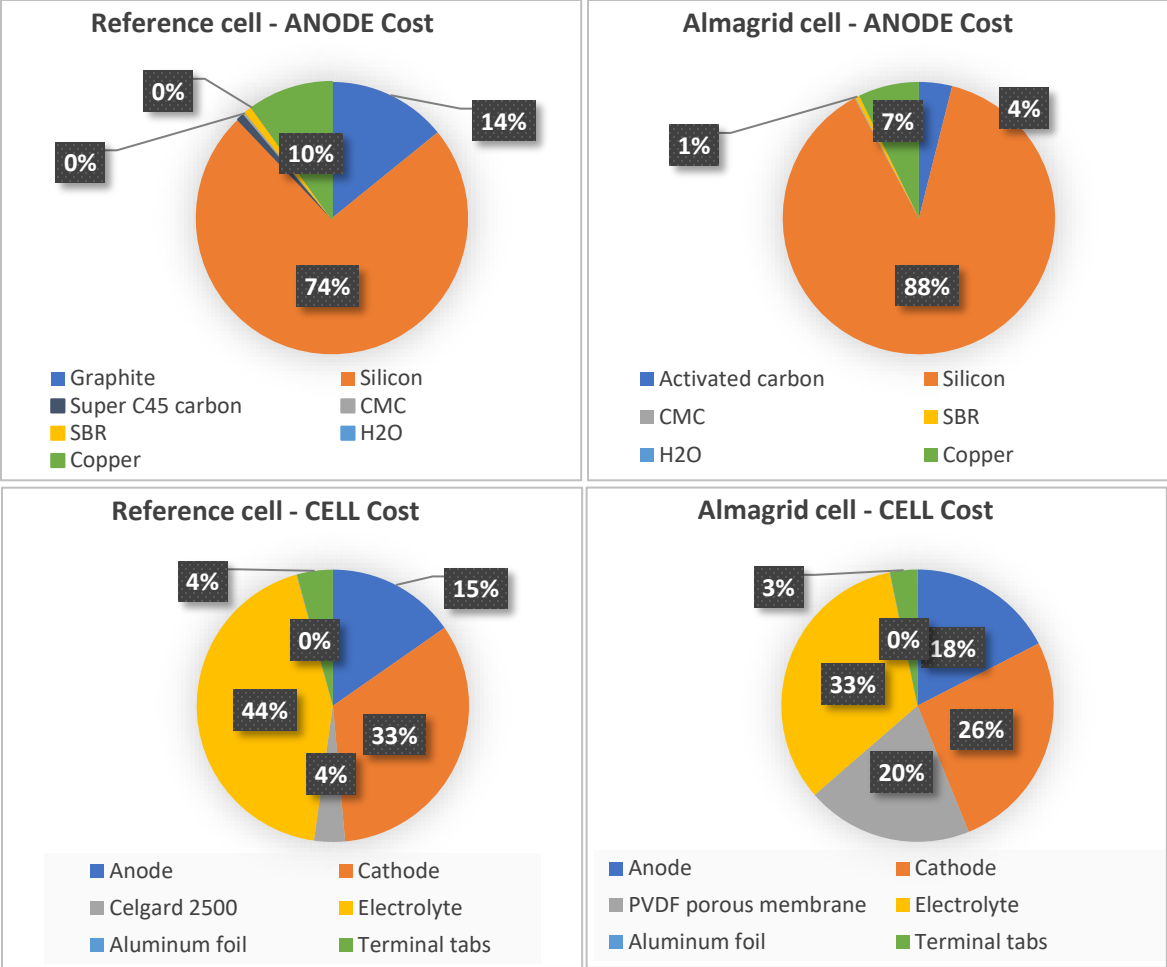


Figure 7. Breakdown of the raw material costs for Reference and Almagrid anode, cathode, and cells. Source: LCC.

3.2.3. Energy cost

Table S. 9 from the SI shows the energy consumption of each piece of equipment involved in the fabrication of the lithium-ion cells. As can be seen in the table, the consumption depends on the type of equipment and the amount of processed material. **Table 8** shows the total consumption of the processes. The anode and cathode manufacturing can be compared, as they are defined by the same units. However, the energy consumption of the cell assembly and the cell finishing processes is given by kWh per cell, so can't be compared to the active materials manufacturing processes. In terms of active materials manufacturing, it is seen that cathode manufacturing requires more energy than the anode. This is mainly due to the higher power requirements of the planetary mixer and longer processing times that can be found in cathode manufacturing. The longer processing times apply in the processes of slurry preparation, casting and drying, due to the higher mass of cathode (Reference: 387.00g; Almagrid: 403.56g) per cell compared to the anode one (Reference: 147.70g; Almagrid: 96.99g). Specially for the casting and the drying processes, much more mass must be coated and dried on the same current collector surface for the cathode manufacturing, leading to a higher cathode thickness and to this increase in power requirements and processing times. In the case of the cell assembly and the cell finishing, similar energy consumptions are found. However, the vacuum drying of the cell involves 67% of the total energy

consumption for the cell assembly process. For the cell finishing process, the highest energy demand process is the aging/grading test, which accounts for the 88% of the energy. **Figure S. 1 from the supplementary information** shows the breakdown of the energy consumption of the different manufacturing processes.

Table 8. Energy consumption for the different manufacturing processes. Source: LCC.

Stage	Energy consumption	Unit
1-A - Anode manufacturing	84.34	kWh/kg
1-B - Cathode manufacturing	155.70	kWh/kg
2 - Cell assembly	21.29	kWh/cell
3 - Cell finishing	18.91	kWh/cell

Table 9 shows the annual energy consumptions cost per process for the production of the reference and Almagrid cells, as well as the total cost. As can be seen, the anode manufacturing is the same for both types of cells, as the same amount of anode (1,000g / 1 batch) is produced per day. The cathode manufacturing stage is more energy-demanding, and expensive, in the case of the Almagrid cell. This is due to the lower amount of anode material needed by the Almagrid cell, which provokes a higher demand for cathode material. Due to the same reason, more Almagrid cells can be produced per day, in comparison with the reference cell (10 Almagrid cells vs 7 reference cells), which leads to the higher annual cost for the cell assembly and cell finishing processes for this type of cell. In result, Almagrid cell production involves an increase of 35% of the energy cost in comparison with the production of the reference cells. However, the energy consumption per produced cell is 9€/cell for both types of cells, which implies that the type of cell does not affect the efficiency of the process in terms of energy cost.

Table 9. Annual energy consumption costs for the Reference and Almagrid cells. Source: LCC.

Stage	Energy consumption cost (€)	
	Reference cell	Almagrid cell
1-A - Anode manufacturing (€)	2,227	2,227
1-B - Cathode manufacturing (€)	10,770	17,103
2 - Cell assembly (€)	3,805	5,795
3 - Cell finishing (€)	2,960	5,148
Total (€)	19,762	30,273
Energy consumption cost per cell (cell/year)		
Cell production	2234	3402
Cost per cell	9	9

Figure S. 2 from the SI shows the breakdown of the annual energy consumption costs related to the manufacturing processes of the reference and Almagrid cells. As was expected, the cathode manufacturing stage is responsible for more than the 50% of the energy consumption. This percentage is a little bit higher in the case of the Almagrid cell (57% vs. 53% for the Reference cell) due to the higher

amount of cathode produced. Nevertheless, there are not significant differences among the manufacturing processes percentages for both cells, as the production stages are the same.

3.2.4. Labour costs

Table 10 shows the C_L needed for each manufacturing stage. In this case, there are no differences regarding the manufacturing of the Reference or Almagrid cells. For the manufacturing of the anode the same amount of material is produced for both cells, so the same labor resources are spent. The cathode manufacturing also employs the same labor resources for both types of cells, even though the amount of cathode manufactured for the Almagrid cell is higher. As was explained before, the planetary mixer has enough capacity to supply the daily production for both types of cells in one batch. This implies that just one operator is in charge of this process per shift in both cases. In the case of the cell assembly, this line is semi-automatic, so one operator is needed for its well-functioning, and its capacity is higher than 10 cells per day, broadly covering the demand for the reference or the Almagrid cells production process. The same happens for the cell finishing process. The most labor-demanding step of this stage is the connection and disconnection of the cells to the electrical testing bench, and no differences in processing times are found when around 6 or 10 cells are connected (daily production of reference and Almagrid cells). However, due to the higher number of cells produced in the case of the Almagrid cell, the C_L per cell is lower comparison with the C_L for the Reference cells production.

Table 10. Annual C_L costs of the manufacturing plant. Source: LCC.

Stage	Operators per day	Personnel time (h)	C_L (€/year)
1-A - Anode manufacturing	2	16	51,163
1-B - Cathode manufacturing	2	16	51,163
2 - Cell assembly	1	8	25,582
3 - Cell finishing	1	8	25,582
Total	6	48	153,490
C_L per cell (€/cell)			
Reference cell		68.70	
Almagrid cell		45.11	

3.2.5. Waste costs

All production processes involve the generation of scrap due to production issues as no production process is 100% efficient. In this way, just one spot of scrap is ideally identified during the manufacturing process of these lithium-ion cells. 5% of scrap is generated during the cell assembly process. The cost of disposal of this scrap is calculated based on the transport of the waste and the waste cost fixed by the waste managers. This waste is composed by the anode material, separator, cathode material, aluminum foil, and copper tabs. The electrolyte scrap is supposed to evaporate during the cell assembly. **Table 11** shows the annual C_W due to the production of the cells. This cost is directly linked to the total amount of material spent by year. In the case of the Almagrid cell, due to the higher production, the C_W due to the disposal of all the components apart from the anode are higher than in the case of the Reference cell. Furthermore, the C_W generated by the separator scrap is much higher in the case of the Almagrid cells. Th

reason behind is the higher amount of separator used in this cell, as the final weight of this separator in the cell is 78.04 g, in contrast with the 17.91 g of separator employed for the production of one Reference cell. There is an exception related to the anode C_w , as this cost is higher in the case of the reference cell. This is because of the lower amount of anode material used in the Almagrid cells, which implies an anode production lower than the one found for the Reference cells, and thus a lower C_w . As happened with other costs, the total C_w is higher for the Almagrid production, but the C_w per cell is very similar for both types of cells. More details about the C_w can be found in **Table S. 13 and Table S. 14 from the SI**.

Table 11. Annual C_w of the generated scrap during the manufacturing process of the Reference and the Almagrid cells. Source: LCC.

Component	Total C_w (€/year)	
	Reference cell	Almagrid cell
Anode	680.21	546.43
Cathode	1,782.26	2,273.67
Separator	323.76	1,218.65
Aluminum foil	78.94	96.57
Terminal tabs	25.33	30.99
Total	2,890.49	4,166.31
C_w per cell (€/cell)		
Reference cell	1.29	
Almagrid cell	1.22	

3.2.6. Externalities costs

Table S. 15 from the SI shows the externalities' costs associated with the production and transport of the Reference and Almagrid cells. To analyze these costs, it is relevant to consider the cost of each of the environmental impacts reflected in the **Table 3**. For example, the most expensive EI is the Particulate matter formation (65 €/kg PM_{2.5} eq); this explains the highest cost of the PM even though the EI was not the highest one. Something similar happens with the Terrestrial acidification, that has a high externality cost (6 €/kg SO₂ eq), but a low impact (around 0.4 kg SO₂ eq/cell). Apart from PM and TA, Human non-carcinogenic toxicity, Climate change, and Ionising radiation were the most expensive environmental impacts due to their higher value due to the LCA results (see **Figure 8**). In this figure, it can be seen that the externalities cost due to the Reference and Almagrid cell is very similar, as the EIs values only differ in a maximum of 5% (the same difference found in the LCA).

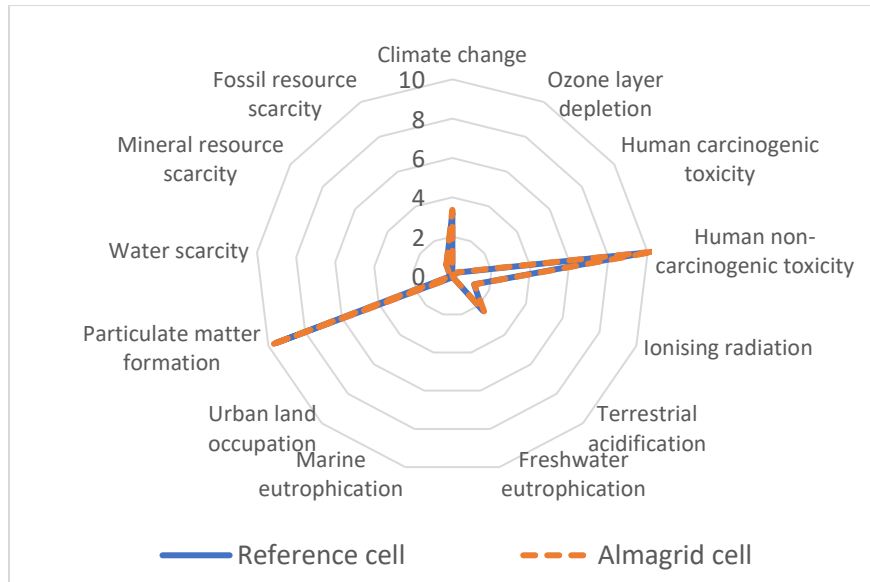


Figure 8. Externalities costs for Reference and Almagrid cells (€/cell). Source: LCC.

3.2.7. Final costs

Table 12 shows the final data related to the cost of the single cells as well as the annual production cost (CAPEX/year). This last value has been already analyzed, and directly depends on the number of produced cells, therefore, the annual cost due to the Reference cell production is much lower (-41%) due to the lower amount of cells production (-34%) in comparison with the Almagrid one. It is nevertheless important to remark this difference is not only caused by the cell production, as raw materials cost is much higher for the Almagrid cell due to the use of the innovative separator with higher thickness and weight, more expensive than graphite activated carbon, and the higher amount of silicon of this cell. It can be seen also that the rest of the costs are very similar for both cells and the annual production, differing by around 30% for the Reference cell in the case of the annual production and in less than 10% for the single cells' values. Considering these facts, the main driver of the cost is the raw materials cost, which accounts 73% of the cost for the Almagrid cell and 62% for the Reference one. The cost of the kWh results in 1.56 €/kWh and 1.73 €/kWh for the Reference and the Almagrid cell respectively, accounting the same differences as the ones found for the cell cost due to both cells storing the same amount of energy.

Table 12. Total cost for the Reference and Almagrid annual production and single cell. Source: LCC.

Cell	Reference		Almagrid	
	Annual cost (k€/year)	Cell cost (€/cell)	Annual cost (k€/year)	Cell cost (€/cell)
Raw materials	391	175	782	230
Labor	153	69	153	45
Electricity	20	9	30	9
Waste	3	1	4	1
Externalities	62	28	98	29
Total	629	282	1068	314

The raw materials identified as cost drivers in both cells are the electrolyte, the LNMO, and the silicon. All these components are mainly made of critical raw materials (CRM) such as lithium, and silicon as was specified in 2020 in (51). However, manganese and nickel are very close to becoming CRM due to their economic importance. The future cell cost is difficult to project due to the use of these CRMs that can suffer shortages in the coming years because of the increase in LIB demand and because of their use in other renewable sectors too, such as photovoltaics. In addition, the boost of the LIB recycling value chain will also determine the future cost of these raw materials. Two conclusions can be extracted from the cost drivers: (1) an increase in the electricity cost will not determine the cost of the cells, and (2) the number of reference cells produced by year must be increased to reduce the final cost of this cell type.

4. Conclusions

In this study, two lithium-ion cells based on LNMO cathode for stationary application have been compared in environmental and economic terms. Reference cell anode contains 10% of silicon and 74% of graphite, in contrast with the Almagrid cell which contains 50% of silicon and 30% of activated carbon. All these changes involve a reduction by 34% of the weight of the Almagrid anode. In addition, Almagrid cell employs a PVDF based porous membrane whose wettability allows the immobilization of the electrolyte increasing the safety of the cells during operation.

Life Cycle Assessment is focused on cells development with special attention to the anodes. Although different impacts considered in Recipe method are analysed, special interest is given to global warming impact. In the case of the cells, Reference overall impact is 49.5 kg CO₂ equivalent whereas for Almagrid cell the value obtained is 51.7 kg CO₂ equivalent. Trends observed in terms of impacts breakdown are similar being cathode, electricity and anode the main contributors. Especially noticeable is the case of the cathode, accounting for about 60% of the total cell impact. Highest contribution to cathode impact is assigned to electricity since its production entails high energy demand.

As for the anode, it should be noted that Reference anode total impact in terms of global warming indicator is 37 kg CO₂ equivalent per kg of anode, while Almagrid anode impact is 53.9 kg CO₂ equivalent per kg of anode. Difference observed is noticeable but given the higher capacity of Almagrid anode this difference is offset since smaller amount of anode is needed for cell integration. Trend observed is the same, being electricity employed in the production process main contributor to total impact in both anodes. Electricity is highly dependent on the country's electricity mix considered and in this study energy mix considered includes 50% coal-based energy. The copper impact is also remarkable since it represents 20-25% due to the highly energy-intensive process of copper mining.

The LCC results are given in terms of raw materials, labor, energy, waste, and externalities costs. Total costs are 282€/cell for the Reference cell and 314€/cell for the Almagrid one. The main driver of the cell costs is the raw materials costs, accounting for 73% of the cost for the Almagrid cell and 62% for the Reference one. Raw materials for Almagrid cell are 31% more expensive due mainly to the higher concentration of silicon, and the higher cost of the activated carbon in contrast with the graphite. In addition, the use in the Almagrid cell of an innovative PVDF porous membrane as a separator increases the cost of the separator by 7 times (45.41€ vs. 6.41€). Labor cost is the same for both cells, nevertheless, in terms of percentages, 24% of the cost of the Reference cell is the labor cost, in contrast with the 12%

spent in labor cost for the Almagrid cell production. The reason behind this is the lower anode mass used in the Almagrid cell, which allows the production of more cells per day compared to the Reference cell production (10 cells vs. 7 cells per day). Energy and waste costs vary along both cells due to the production cell amounts. In the case of the externalities cost, Particulate matter formation, terrestrial acidification, Human non-carcinogenic toxicity, Climate change, and Ionising radiation are the most expensive environmental impacts according to the LCA results, and the higher External Cost Factors of Particulate matter formation and terrestrial acidification. The kWh costs are 1.56 €/kWh and 1.73 €/kWh for the Reference and the Almagrid cell respectively, indicating a higher cost for the Almagrid cell.

As a summary, the results show that the Almagrid cell is expected to reduce the environmental impact due to the anode mass reduction while fulfilling the same electrical performance as the Reference cell and showing better safety under operation thanks to the PVDF porous membrane. Although results obtained for both cells are similar, it should be noted that cell and components (anodes and Almagrid separator) have been developed at laboratory scale and their optimization and scaling up are expected to reduce current impacts and process costs.

5. Outlook

Based on the article's conclusions, the focus of the new generation of LNMO batteries with gel polymer electrolytes and silicon anodes will primarily be on optimizing and upscaling the cell components. One of the main points to address will be reducing the separator thickness which would result in an approximate 50% decrease in separator weight. Consequently, optimizing the separator will contribute to a reduction in the environmental impact of the Almagrid cell and, subsequently, its price.

In the case of the carbon footprint a reduction of 50% in separator's mass would turn out in a reduction of 1.4% of the global warming indicator value. Almagrid cell overall impact is 51.7 kg CO₂ equivalent whereas for the optimized Almagrid cell the value obtained is 51 kg CO₂ equivalent. Carbon footprint of this cell is 3% higher than Reference cell.

Regarding the cell cost, reducing the separator thickness would result in a 10% decrease in cell cost, amounting to 291€/cell compared to 314€/cell without separator reduction. This reduction is achieved by saving 10% less raw materials expenditure when the separator weight is halved. In comparison to the Reference cell, the optimized Almagrid cell may be slightly more expensive, around 3% higher.

In conclusion and according to the low Technology Readiness Level (around 5) of the Almagrid cell, the optimization and industrialization of the different components will lead to environmental impact and cost reduction. For this reason, the development of these new components is essential and should continue to support the new challenges considered in the Green Deal regarding the grid-storage and electromobility.

6. References

1. EUROSTAT. Renewable energy statistics [Internet]. [cited 2023 Jun 1]. Available from: <https://ec.europa.eu/eurostat/statistics->

explained/index.php?title=Renewable_energy_statistics#Share_of_renewable_energy_more_than_doubled_between_2004_and_2020

2. ALTENEX energy. The Consequences of the European Energy Crisis Amid the Global Pandemic [Internet]. [cited 2023 Jun 1]. Available from: <https://www.altenexenergy.com/news/the-consequences-of-the-european-energy-crisis-amid-the-global-pandemic/>
3. European Environment Energy. Trends and projections: limited rebound in EU emissions amid post-pandemic recovery and energy crisis [Internet]. [cited 2023 Jun 1]. Available from: <https://www.eea.europa.eu/highlights/trends-and-projections-limited-rebound>
4. Dunn B, Kamath H, Tarascon JM. Electrical Energy Storage for the Grid: A Battery of Choices. *Science* (1979) [Internet]. 2011;334(6058):928–35. Available from: <https://www.science.org/doi/abs/10.1126/science.1212741>
5. Palizban O, Kauhaniemi K. Energy storage systems in modern grids—Matrix of technologies and applications. *J Energy Storage* [Internet]. 2016;6:248–59. Available from: <https://www.sciencedirect.com/science/article/pii/S2352152X1630010X>
6. Claus Daniel (Editor), J. O. Besenhard (Editor). *Handbook of Battery Materials*, 2nd Edition [Internet]. 2012 [cited 2023 Jun 1]. 1–1023 p. Available from: <https://books.google.es/books?hl=es&lr=&id=mHhSqlL1TeoC&oi=fnd&pg=PT10&dq=J.O.+Handbook+of+Battery+Materials,+2nd+ed.%3B+Wiley:+Weinheim,+Germany,+2012.&ots=LFNg7sF2ge&sig=tUShHu7HKz6TzWFNgFpYjUYYP3I#v=onepage&q&f=false>
7. Wood Mackenzie. LFP to overtake NMC as dominant stationary storage chemistry by 2030 [Internet]. 2020 [cited 2023 Jun 1]. Available from: <https://www.woodmac.com/press-releases/lfp-to-overtake-nmc-as-dominant-stationary-storage-chemistry-by-2030/>
8. Amlı H, Booth M, Dhanak V, Ahmed W. Chapter 19 - Recent developments in battery technologies. In: Ahmed W, Booth M, Nourafkan E, editors. *Emerging Nanotechnologies for Renewable Energy* [Internet]. Elsevier; 2021. p. 517–43. (Micro and Nano Technologies). Available from: <https://www.sciencedirect.com/science/article/pii/B978012821346900002X>
9. TOPSOE. THE CATHODE MATERIAL FOR NEXT-GENERATION LITHIUM ION BATTERIES IS READY [Internet]. 2019 [cited 2023 Jun 1]. Available from: <https://www.topsoe.com/blog/the-cathode-material-for-next-generation-lithium-ion-batteries-is-ready>
10. Amin R, Muralidharan N, Petla RK, Ben Yahia H, Jassim Al-Hail SA, Essehli R, et al. Research advances on cobalt-free cathodes for Li-ion batteries - The high voltage LiMn_{1.5}Ni_{0.5}O₄ as an example. *J Power Sources* [Internet]. 2020;467:228318. Available from: <https://www.sciencedirect.com/science/article/pii/S0378775320306224>
11. Li P, Kim H, Myung ST, Sun YK. Diverting Exploration of Silicon Anode into Practical Way: A Review Focused on Silicon-Graphite Composite for Lithium Ion Batteries. *Energy Storage*

- Mater [Internet]. 2021;35:550–76. Available from:
<https://www.sciencedirect.com/science/article/pii/S2405829720304384>
12. Kristina Edström. BATTERY 2030+ Roadmap - INVENTING THE SUSTAINABLE BATTERIES OF THE FUTURE BATTERY 2030+ [Internet]. [cited 2023 Jun 1]. Available from:
https://battery2030.eu/wp-content/uploads/2022/07/BATTERY-2030-Roadmap_Revision_FINAL.pdf
 13. Zuo X, Zhu J, Müller-Buschbaum P, Cheng YJ. Silicon based lithium-ion battery anodes: A chronicle perspective review. *Nano Energy* [Internet]. 2017;31:113–43. Available from:
<https://www.sciencedirect.com/science/article/pii/S2211285516304931>
 14. Nangir M, Massoudi A, Tayebifard SA. Investigation of the lithium-ion depletion in the silicon-silicon carbide anode/electrolyte interface in lithium-ion battery via electrochemical impedance spectroscopy. *Journal of Electroanalytical Chemistry* [Internet]. 2020;873:114385. Available from:
<https://www.sciencedirect.com/science/article/pii/S1572665720306123>
 15. Engels P, Cerdas F, Dettmer T, Frey C, Hentschel J, Herrmann C, et al. Life cycle assessment of natural graphite production for lithium-ion battery anodes based on industrial primary data. *J Clean Prod* [Internet]. 2022;336:130474. Available from:
<https://www.sciencedirect.com/science/article/pii/S0959652622001172>
 16. Jiang J, Zhu J, Ai W, Fan Z, Shen X, Zou C, et al. Evolution of disposable bamboo chopsticks into uniform carbon fibers: a smart strategy to fabricate sustainable anodes for Li-ion batteries. *Energy Environ Sci* [Internet]. 2014;7(8):2670–9. Available from:
<http://dx.doi.org/10.1039/C4EE00602J>
 17. Muraleedharan Pillai M, Kalidas N, Zhao X, Lehto VP. Biomass-Based Silicon and Carbon for Lithium-Ion Battery Anodes. *Front Chem* [Internet]. 2022;10. Available from:
<https://www.frontiersin.org/articles/10.3389/fchem.2022.882081>
 18. Jiang J, Zhu J, Ai W, Fan Z, Shen X, Zou C, et al. Evolution of disposable bamboo chopsticks into uniform carbon fibers: a smart strategy to fabricate sustainable anodes for Li-ion batteries. *Energy Environ Sci* [Internet]. 2014;7(8):2670–9. Available from:
<http://dx.doi.org/10.1039/C4EE00602J>
 19. Sbrascini L, Staffolani A, Bottoni L, Darjazi H, Minnetti L, Minicucci M, et al. Structural and Interfacial Characterization of a Sustainable Si/Hard Carbon Composite Anode for Lithium-Ion Batteries. *ACS Appl Mater Interfaces* [Internet]. 2022 Jul 27;14(29):33257–73. Available from: <https://doi.org/10.1021/acsami.2c07888>
 20. Adams RA, Dysart AD, Esparza R, Acuña S, Joshi SR, Cox A, et al. Superior Lithium-Ion Storage at Room and Elevated Temperature in an Industrial Woodchip Derived Porous Carbon. *Ind Eng Chem Res* [Internet]. 2016 Aug 17;55(32):8706–12. Available from:
<https://doi.org/10.1021/acs.iecr.6b01786>
 21. Xiang J, Lv W, Mu C, Zhao J, Wang B. Activated hard carbon from orange peel for lithium/sodium ion battery anode with long cycle life. *J Alloys Compd* [Internet].

2017;701:870–4. Available from:

<https://www.sciencedirect.com/science/article/pii/S0925838817302402>

22. Yokokura TJ, Rodriguez JR, Pol VG. Waste Biomass-Derived Carbon Anode for Enhanced Lithium Storage. *ACS Omega* [Internet]. 2020 Aug 11;5(31):19715–20. Available from: <https://doi.org/10.1021/acsomega.0c02389>
23. Wang H, Zhang P, Song X, Zhang M, Kong X, Jin S, et al. Wheat Bran Derived Carbon toward Cost-Efficient and High Performance Lithium Storage. *ACS Sustain Chem Eng* [Internet]. 2020 Oct 26;8(42):15898–905. Available from: <https://doi.org/10.1021/acssuschemeng.0c04670>
24. Lee H, Yanilmaz M, Toprakci O, Fu K, Zhang X. A review of recent developments in membrane separators for rechargeable lithium-ion batteries. *Energy Environ Sci* [Internet]. 2014;7(12):3857–86. Available from: <http://dx.doi.org/10.1039/C4EE01432D>
25. Li A, Yuen ACY, Wang W, De Cachinho Cordeiro IM, Wang C, Chen TBY, et al. A Review on Lithium-Ion Battery Separators towards Enhanced Safety Performances and Modelling Approaches. *Molecules* [Internet]. 2021;26(2). Available from: <https://www.mdpi.com/1420-3049/26/2/478>
26. Lu W, Yuan Z, Zhao Y, Zhang H, Zhang H, Li X. Porous membranes in secondary battery technologies. *Chem Soc Rev* [Internet]. 2017;46(8):2199–236. Available from: <http://dx.doi.org/10.1039/C6CS00823B>
27. Wu Y, Li Y, Wang Y, Liu Q, Chen Q, Chen M. Advances and prospects of PVDF based polymer electrolytes. *Journal of Energy Chemistry* [Internet]. 2022;64:62–84. Available from: <https://www.sciencedirect.com/science/article/pii/S2095495621002096>
28. Zhou D, Shanmukaraj D, Tkacheva A, Armand M, Wang G. Polymer Electrolytes for Lithium-Based Batteries: Advances and Prospects. *Chem* [Internet]. 2019;5(9):2326–52. Available from: <https://www.sciencedirect.com/science/article/pii/S2451929419302190>
29. Zhu M, Wu J, Wang Y, Song M, Long L, Siyal SH, et al. Recent advances in gel polymer electrolyte for high-performance lithium batteries. *Journal of Energy Chemistry* [Internet]. 2019;37:126–42. Available from: <https://www.sciencedirect.com/science/article/pii/S2095495618309331>
30. Arshad F, Lin J, Manurkar N, Fan E, Ahmad A, Tariq M un N, et al. Life Cycle Assessment of Lithium-ion Batteries: A Critical Review. *Resour Conserv Recycl* [Internet]. 2022;180:106164. Available from: <https://www.sciencedirect.com/science/article/pii/S092134492200012X>
31. Commission E, Centre JR, Cristobal-Garcia J, Pant R, Reale F, Sala S. Life cycle assessment for the impact assessment of policies. Publications Office; 2017.
32. Porzio J, Scown CD. Life-Cycle Assessment Considerations for Batteries and Battery Materials. *Adv Energy Mater* [Internet]. 2021;11(33):2100771. Available from: <https://onlinelibrary.wiley.com/doi/abs/10.1002/aenm.202100771>

33. Pellow MA, Ambrose H, Mulvaney D, Betita R, Shaw S. Research gaps in environmental life cycle assessments of lithium ion batteries for grid-scale stationary energy storage systems: End-of-life options and other issues. *Sustainable Materials and Technologies* [Internet]. 2020;23:e00120. Available from: <https://www.sciencedirect.com/science/article/pii/S2214993718302318>
34. Peters JF, Weil M. Providing a common base for life cycle assessments of Li-Ion batteries. *J Clean Prod* [Internet]. 2018;171:704–13. Available from: <https://www.sciencedirect.com/science/article/pii/S0959652617323077>
35. Hiremath M, Derendorf K, Vogt T. Comparative Life Cycle Assessment of Battery Storage Systems for Stationary Applications. *Environ Sci Technol* [Internet]. 2015 Apr 21;49(8):4825–33. Available from: <https://doi.org/10.1021/es504572q>
36. Working Group 6. ROADMAP ON STATIONARY APPLICATIONS FOR BATTERIES [Internet]. 2022 [cited 2023 Jun 1]. Available from: <https://energy.ec.europa.eu/system/files/2022-01/vol-6-009.pdf>
37. BloombergNEF. Lithium-ion Battery Pack Prices Rise for First Time to an Average of \$151/kWh [Internet]. 2022 [cited 2023 Jun 8]. Available from: <https://about.bnef.com/blog/lithium-ion-battery-pack-prices-rise-for-first-time-to-an-average-of-151-kwh/>
38. Rahman MM, Oni AO, Gemechu E, Kumar A. Assessment of energy storage technologies: A review. *Energy Convers Manag* [Internet]. 2020;223:113295. Available from: <https://www.sciencedirect.com/science/article/pii/S0196890420308347>
39. Hesse HC, Schimpe M, Kucevic D, Jossen A. Lithium-Ion Battery Storage for the Grid—A Review of Stationary Battery Storage System Design Tailored for Applications in Modern Power Grids. *Energies (Basel)* [Internet]. 2017;10(12). Available from: <https://www.mdpi.com/1996-1073/10/12/2107>
40. Zakeri B, Syri S. Electrical energy storage systems: A comparative life cycle cost analysis. *Renewable and Sustainable Energy Reviews* [Internet]. 2015;42:569–96. Available from: <https://www.sciencedirect.com/science/article/pii/S1364032114008284>
41. Díaz-Ramírez MC, Blecua-de-Pedro M, Arnal AJ, Post J. Acid/base flow battery environmental and economic performance based on its potential service to renewables support. *J Clean Prod* [Internet]. 2022;330:129529. Available from: <https://www.sciencedirect.com/science/article/pii/S0959652621037082>
42. Landa-Medrano I, Eguia-Barrio A, Sananes-Israel S, Porcher W, Trad K, Moretti A, et al. Insights into the Electrochemical Performance of 1.8 Ah Pouch and 18650 Cylindrical NMC:LFP|Si:C Blend Li-ion Cells. *Batteries* [Internet]. 2022;8(8). Available from: <https://www.mdpi.com/2313-0105/8/8/97>
43. Indeed [Internet]. [cited 2022 Dec 1]. Available from: https://es.indeed.com/career/operario-producci%C3%B3n/salaries/Donostia~San-Sebasti%C3%A1n--Guip%C3%BAcoa-provincia?from=top_sb

44. Yves Gérard, Stéphane Le Pochat, Anaëlle Dubosc. Modélisation des externalités environnementales pour une TVA circulaire (MODEXT) – Rapport final, Ademe [Internet]. 2018 [cited 2023 Jan 12]. Available from: http://www.fondation-2019.fr/wp-content/uploads/2019/06/MODEXT_Rapport-final_ADEME_vf2_190604.pdf
45. EUROSTAT Data Browser [Internet]. [cited 2023 Jan 12]. Available from: https://ec.europa.eu/eurostat/databrowser/view/PRC_HICP_AIND/default/table?lang=en&category=prc.prc_hicp
46. Instituto Nacional de Estadística (INE) [Internet]. [cited 2023 Jan 1]. Available from: https://www.ine.es/prensa/ipca_prensa.htm
47. Sander de Bruyn, Marijn Bijleveld, Lonneke de Graaff, Ellen Schep, Arno Schroten, Robert Vergeer, et al. 7.N54 – Environmental Prices Handbook [Internet]. 2018 [cited 2023 Jan 12]. Available from: https://cedelft.eu/wp-content/uploads/sites/2/2021/04/CE_Delft_7N54_Environmental_Prices_Handbook_EU28_version_Def_VS2020.pdf
48. Yadav P, Ismail N, Essalhi M, Tysklind M, Athanassiadis D, Tavajohi N. Assessment of the environmental impact of polymeric membrane production. J Memb Sci [Internet]. 2021;622:118987. Available from: <https://www.sciencedirect.com/science/article/pii/S037673882031560X>
49. Danny Wille. Annex: Monetisation of the MMG method [Internet]. 2017 [cited 2023 Jan 12]. Available from: <https://publicaties.vlaanderen.be/view-file/26888>
50. Amadei AM, De Laurentiis V, Sala S. A review of monetary valuation in life cycle assessment: State of the art and future needs. J Clean Prod [Internet]. 2021;329:129668. Available from: <https://www.sciencedirect.com/science/article/pii/S0959652621038452>
51. Study on the EU's List of Critical Raw Materials – Final Report (2020). 2020.

Aknowlegments:

This work was supported by the RED CERVERA Almagrid (CER-20191006) “Integración de tecnologías avanzadas de almacenamiento de energía para aplicaciones de red” that has received funding from the Centro para el Desarrollo Tecnológico e Industrial (CDTI) from the Science and Innovation Ministry from Spain.

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