



Data Article

ESCAPE approach for the sustainability evaluation of spent lithium-ion batteries recovery: Dataset of 33 available technologies

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ABSTRACT

Recovering critical raw materials from end-of-life batteries is mandatory to limit the need of virgin resources in the long-term. However, most of the recycling of lithium-ion batteries (LIBs) technologies are still in an infancy stage. As a result, to date, only few studies focus on Life Cycle Assessment (LCA) of the proposed processes, presenting limited results.

This paper reports the methodology and data resulting from sustainability evaluation of 33 different technologies for spent LIBs recovery, on the basis of the availability of information, identified in literature. The ESCAPE (standing for Evaluation of Sustainability of material substitution using Carbon footPrint by a simplified approach) method is based on the use of only two parameters: the embodied energy and the carbon footprint. These parameters are calculated for all the process steps of each technology. Using the ESCAPE approach, the data about energies and emissions associated with the electricity consumption for thermal and mechanical treatments and chemicals and water use are calculated for

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all the 33 selected technologies, referring to a recent work (Fahimi et al., 2022), which only presents the results. In addition, ESCAPE tool is used to evaluate and discuss the parameters that can affect the technologies sustainability, to better highlight the most onerous and impactful steps of each technology. Then, this paper also shows that ESCAPE approach allows to propose some strategies to improve the recovery processes, with the aim to support eco-design.

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Specifications Table

| | |
|--------------------------------|--|
| Subject | Environmental science (General) |
| Specific subject area | Sustainability evaluation of raw materials recovery from spent lithium-ion batteries, based on embodied energy and carbon footprint |
| Type of data | Table Figure |
| How the data were acquired | Data were elaborated using the approach presented in this paper |
| Data format | Analyzed |
| Description of data collection | Referring to laboratory scale, 33 available technologies for LIBs recovery were analyzed. Every process was divided in single steps (considering chemicals, water, thermal and mechanical treatments) to calculate embodied energy and carbon footprint and, if possible, compared to reference material (extracted from virgin source). Data were referred to 1 kg cathode. |
| Data source location | Data evaluated following the procedures reported in ref [1]. are available in this work. |
| Data accessibility | With the article |
| Related research article | A. Fahimi, S. Ducoli, S. Federici, G. Ye, E. Mousa, P. Frontera, E. Bontempi, Evaluation of the sustainability of technologies to recycle spent lithium-ion batteries, based on embodied energy and carbon footprint, J. Clean. Prod, 338 (2022) 130493. https://doi.org/10.1016/j.jclepro.2022.130493 |

Value of the Data

- Several studies have proposed the possibility to recover a variety of materials from LIBs, even if the processes are developed only at the lab-scale.
- The ESCAPE approach is presented and applied to evaluate the available strategies to recover materials from LIBs.
- Following this approach, the data about energy consumption and emissions are calculated for all the steps of the 33 selected literature processes, proposed for LIBs recovery.
- The calculated data are used to evaluate the sustainability of selected technologies, allowing to provide an instrument to support the most suitable activities able to extract materials from waste acting in substitution of natural resources use.
- The parameters evaluated in the ESCAPE approach (embodied energy and carbon footprint) can be potentially integrated and/or compared with Life Cycle Assessment (LCA) study, giving an initial overview of a process even if developed at low technology readiness level.
- ESCAPE approach allows also to propose eco-design strategies for reducing environmental impact. For example, this work shows that water usage must be suitably managed (for example, limiting the use of ultrapure water) to improve the sustainability of LIBs recycling technologies.

1. Data Description

Table 1 reports conditions and parameters used for the evaluation of the embodied energy and the carbon footprint of the processes. They include also hypotheses whenever it is required

to set missing information in the referring literature source (e.g. washing step described without mentioning any volume and type of water).

Table 2 reports the average power rating of laboratory instruments used for laboratory scale processes for treating 1 kg of material.

Table 3 reports the embodied energy and the carbon footprint values of chemicals used in this work and extracted from CES Selector (<https://grantadesign.com/it/industry/products/ces-selector/>) or Ecoinvent database (<https://ecoinvent.org/>).

Table 4 reports the embodied energy and the carbon footprint referred to a power rating of 1 W and for 60 s of usage, considered as global “World factors”, as average value of all world countries. The data were calculated by considering the reports of the International Energy Agency (IEA) [2,3,5].

Table 5 reports the detailed values of the embodied energy and the carbon footprint evaluated for all the steps of each considered process (see also Ref [1]), divided into four categories: thermal treatments, mechanical treatments, chemicals, and water use. The data were calculated considering the processes as exactly described by the authors.

Table 6 reports the detailed values of the embodied energy and the carbon footprint evaluated for all the steps of each considered process (see Ref. also [1]), divided into four categories: thermal treatments, mechanical treatments, chemicals, and water use. The data were calculated considering the use of distilled water instead of ultrapure water or deionized water for chemicals dilution. In addition, for the products washing, only tap water was considered.

Table 1

Conditions and parameters used for the evaluation of embodied energy and carbon footprint of the recycling processes for spent lithium-ion batteries.

| | |
|----|--|
| 1 | 1 kg raw material (cathode of batteries) was considered for all the processes. Its embodied energy and carbon footprint are assumed to be equal to zero, since recycling spent LIB is originally considered a waste. |
| 2 | For thermal and mechanical processes, all the available information (about time and temperature) were found in the reference article. |
| 3 | To account the quantities of chemicals, their amount (reported in the reference papers) was adjusted to 1 kg of starting raw material. |
| 4 | For drying processes (made at around 100 °C), a power value of 400 W was used. Time, if not specified, was set to one hour. |
| 5 | For thermal processes at elevated temperature (activation, pyrolysis, carbonization, etc.) a power value of 2500 W was used. |
| 6 | If the reference articles proposed several synthesis conditions, the optimal condition was considered, otherwise if not mentioned, the lowest values of times and/or temperatures were considered (to have the lowest embodied energy and carbon footprint). |
| 7 | In thermal treatments, the additional time required to reach the working temperature (reported only from some authors) has not been considered in the calculations. |
| 8 | When a flow of nitrogen or argon was coupled to the heating, these elements were evaluated as mass of reagent added to the process, and the mass was obtained from their input flow data indicated by the reference article |
| 9 | For mechanical processes, time was set on 5 min, if not specified in the article. |
| 10 | Filtration was considered performed by the operator, without the use of instrument. In any cases this process is expected to have low embodied energy and carbon footprint. |
| 11 | The water used to prepare the solutions of chemicals used in the synthesis was considered distilled. |
| 12 | The water used for washing has been always considered as tap water even if distilled water was expressly indicated. |
| 13 | For each washing step, 10 l of water were considered for 1 kg of starting raw material. |
| 14 | Liquid CO ₂ was evaluated as additional reagent, and whenever not mentioned its flow input, we assumed it to be equal to 5 l/min |
| 15 | For technologies exploiting water leaching, the “leaching” water was considered as tap water. |
| 16 | In case of mechanical/thermal/chemical treatment previous to eventual chemical analysis (e.g. ICP-MS), these were not considered in the calculation. |
| 17 | For drying steps, if temperature not mentioned, we assume it is done under room conditions. |
| 18 | Volume of NaCl (5%) solution for discharging step of batteries was assumed to be 10 l of solution for 1 kg of material. |
| 19 | The efficiency of the processes was not considered in the calculation since in several articles they were not indicated. |

Table 2

List of power rating for thermal and mechanical laboratory processes considered in the work.

| | Process | Power (W) |
|----------------------|---|-----------|
| Thermal processes | Drying (low temperature $\approx 100^{\circ}\text{C}$) | 400 |
| | Heating (high temperature) | 2500 |
| | Heating and mixing | 630 |
| | Laboratory scale autoclave | 3700 |
| | Laboratory scale arc furnace | 2880 |
| | Pilot scale vacuum furnace | 12,000 |
| Mechanical processes | Centrifugation | 500 |
| | Crushing | 1100 |
| | Cutting/Shredding | 1000 |
| | Grinding | 250 |
| | Milling | 1800 |
| | Sieving | 480 |
| | Stirring | 270 |
| | Sonicating | 200 |
| | Vacuuuming | 550 |

Table 3

List of embodied energy and carbon footprint of chemicals used in this work and Ref [1].

| Chemical | Embodied energy (MJ/kg) | Carbon footprint (kgCO ₂ -eq/kg) |
|--|-------------------------|---|
| Citric acid | 74.4 | 3.1 |
| Distilled water | 0.0135 | 0.00082 |
| dH ₂ O (double deionized water) | 19.1 | 0 |
| Tap water | 0.005 | 0.0003 |
| Hydrogen peroxide | 12.94 | 0.01 |
| Gypsum | 0.05 | 0 |
| Hydrochloric acid | 17.5 | 0.9 |
| Isopropyl alcohol | 1.69 | 1.85 |
| Nitrogen (gas) | 4.3 | 0.25 |
| Industrial grade Phosphoric acid | 27.2 | 0.5 |
| Fertilizers grade Phosphoric acid | 18.2 | 1 |
| Sodium bicarbonate | 7.53 | 0.61 |
| Sodium hydroxide | 12.54 | 3.2 |
| Sulfuric acid | 7.36 | 0.21 |
| LiOH | 62.9 | 5.7 |
| Dimethyl carbonate (DMC) | 54.1 | 2.3 |
| Liquid Argon | 32.07 | 2.33 |
| Sodium percarbonate | 18.1 | 1.26 |
| Liquid carbon dioxide | 8.24 | 0.9 |
| Lignite | 9.5 | 0.036 |
| Silica | 33.1 | 3.2 |
| Calcium oxide | 3.7 | 1.2 |
| Dimethyl acetamide (DMAC) | 88.8 | 3.4 |
| Lithium carbonate | 27.24 | 2.06 |
| NaCl | 2.4 | 0.18 |
| EDTA | 78.2 | 4.24 |
| NaCl | 2.4 | 0.18 |
| Ammonium sulphate | 6.2 | 0.5 |
| Nitric acid (50%) | 12.54 | 3.2 |

Table 4

EE and CF referred to a power rating of 1 W and for 1 min of usage, considered as global "World factors", as average value of all world countries. The data were calculated by considering the reports of the International Energy Agency (IEA) [2,3].

| Electric_to_Thermal (1 W; 60 s) | | Electric_to_Mechanical (1 W; 60 s) | |
|---------------------------------|-------------------|------------------------------------|-------------------|
| EE factor (MJ/kg) | CF factor (kg/kg) | EE factor (MJ/kg) | CF factor (kg/kg) |
| 0.00012153 | 0.00000714 | 0.00013656 | 0.00000802 |

Table 5

Detailed values of embodied energy (EE) and carbon footprint (CF) resulted for all the steps of each considered process (see Ref [1]), divided into four categories: thermal treatments, mechanical treatments, chemicals, and water use. (A) hydrometallurgical processes; (B) pyrometallurgical processes; (C) direct recycling processes. The data were calculated considering the processes as exactly described by the authors.

| A: HYDROMETALLURGICAL METHOD | | | | | | | |
|--|---------------|---------------|--|---------------|---------------|--|----------------------------------|
| 1 | | | 2 | | | 3 | |
| https://doi.org/10.1016/j.matchemphys.2017.01.003 | | | DOI: 10.1016/j.jhazmat.2009.11.026 | | | https://doi.org/10.1021/acssuschemeng.7b00571 | |
| <i>Photocatalytic properties of Co3O4/LiCoO2 recycled from spent lithium-ion batteries using citric acid as leaching agent</i> | EE (MJ/kg) | CF (kgCO2/kg) | <i>Recovery of cobalt and lithium from spent lithium ion batteries using organic citric acid as leachant</i> | EE (MJ/kg) | CF (kgCO2/kg) | <i>Sustainable Recovery of Cathode Materials from Spent Lithium-Ion Batteries Using Lactic Acid Leaching System</i> | EE (MJ/kg) CF (kgCO2/kg) |
| Total | 2336,5 | 69,7 | Total | 1899,8 | 48,4 | Total | 1456,6 43,3 |
| Thermal treatment | 160,4 | 9,4 | Thermal treatment | 168,0 | 9,9 | Thermal treatment | 165,6 9,7 |
| Mechanical treatment | 0 | 0 | Mechanical treatment | 4,1 | 0,2 | Mechanical treatment | 1,2 0,1 |
| Chemical treatment | 1441,6 | 60,3 | Chemical treatment | 910,7 | 38,2 | Chemical treatment | 439,3 33,5 |
| Use of water (washing steps and solutions) | 734,5 | 0,0 | Use of water (washing steps and solutions) | 816,9 | 0,0 | Use of water (washing steps and solutions) | 850,5 0,0 |
| 4 | | | 5 | | | 6 | |
| https://doi.org/10.1016/j.wasman.2017.03.037 | | | https://doi.org/10.1016/j.jpowsour.2012.06.068 | | | https://doi.org/10.1016/j.cej.2015.06.071 | |
| <i>Recovery of lithium and cobalt from spent lithium-ion batteries using organic acids: Process optimization and kinetic aspects</i> | EE (MJ/kg) | CF (kgCO2/kg) | <i>Ascorbic-acid-assisted recovery of cobalt and lithium from spent Li-ion batteries</i> | EE (MJ/kg) | CF (kgCO2/kg) | <i>Hydrometallurgical processing of spent lithium ion batteries (LIBs) in the presence of a reducing agent with emphasis on kinetics of leaching</i> | EE (MJ/kg) CF (kgCO2/kg) |
| Total | 1578,5 | 47,7 | Total | 1075,7 | 28,0 | Total | 1018,7 9,3 |
| Thermal treatment | 118,6 | 7,0 | Thermal treatment | 19,8 | 1,2 | Thermal treatment | 97,5 5,7 |
| Mechanical treatment | 0,6 | 0,0 | Mechanical treatment | 1,5 | 0,1 | Mechanical treatment | 0,8 0,0 |
| Chemical treatment | 969,2 | 40,7 | Chemical treatment | 392,3 | 27,8 | Chemical treatment | 118,2 3,6 |

Table 5 (continued)

| | | | | | | | | |
|---|--------------|----------------------------|--|---------------|----------------------------|---|--------------|----------------------------|
| Use of water (washing steps and solutions) | 490,0 | 0,0 | Use of water (washing steps and solutions) | 662,7 | 0,0 | Use of water (washing steps and solutions) | 802,2 | 0,0 |
| 7 | | | 8 | | | 9 | | |
| https://doi.org/10.1016/j.jpowsour.2013.08.128 | | | https://doi.org/10.1016/j.jhazmat.2016.03.062 | | | https://doi.org/10.1016/j.wasman.2016.03.006 | | |
| <i>Hydrometallurgical process for the recovery of high value metals from spent lithium nickel cobalt aluminum oxide based lithium-ion batteries</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Recycling of spent lithium-ion battery cathode materials by ammoniacal leaching</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>An environmental benign process for cobalt and lithium recovery from spent lithium-ion batteries by mechanochemical approach</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 527,2 | 11,3 | Total | 2179,4 | 23,5 | Total | 512,5 | 27,8 |
| Thermal treatment | 155,6 | 9,1 | Thermal treatment | 7,5 | 0,4 | Thermal treatment | 36,5 | 2,1 |
| Mechanical treatment | 1,6 | 0,1 | Mechanical treatment | 1,6 | 0,1 | Mechanical treatment | 75,7 | 4,4 |
| Chemical treatment | 34,4 | 2,0 | Chemical treatment (only the binary ammonia + ammonium sulfite) | 336,7 | 22,9 | Chemical treatment | 390,8 | 21,2 |
| Use of water (washing steps and solutions) | 335,6 | 0,0 | Use of water (washing steps and solutions) | 1833,6 | 0,0 | Use of water (washing steps and solutions) | 9,6 | 0,0 |
| 10 | | | 11 | | | 12 | | |
| https://doi.org/10.1016/j.wasman.2017.05.013 | | | https://doi.org/10.1016/j.wasman.2016.09.039 | | | https://doi.org/10.1016/j.jclepro.2015.10.132 | | |
| <i>Recycling of spent lithium-ion battery with polyvinyl chloride by mechanochemical process</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Green and facile method for the recovery of spent Lithium Nickel Manganese Cobalt Oxide (NMC) based Lithium ion batteries</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>An atom-economic process for the recovery of high value-added metals from spent lithium-ion batteries</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 451,8 | 13,6 | Total | 196,3 | 1,0 | Total | 587,9 | 11,0 |
| Thermal treatment | 42,3 | 2,5 | Thermal treatment | 9,8 | 0,6 | Thermal treatment | 45,6 | 2,7 |
| Mechanical treatment | 147,6 | 8,7 | Mechanical treatment | 1,9 | 0,1 | Mechanical treatment | 0,9 | 0,1 |
| Chemical treatment | 70,4 | 2,4 | Chemical treatment | 4,7 | 0,3 | Chemical treatment | 180,9 | 8,3 |
| Use of water (washing steps and solutions) | 191,5 | 0,0 | Use of water (washing steps and solutions) | 180,0 | 0,0 | Use of water (washing steps and solutions) | 359, | 0,0 |

Table 5 (continued)

| solutions) | | | solutions) | | | steps and solutions) | 8 | |
|--|--------------|----------------------------|--|--------------|----------------------------|---|--------------|----------------------------|
| 13 | | | 14 | | | 15 | | |
| https://doi.org/10.1016/j.wasmat.2016.03.009 | | | https://doi.org/10.1016/j.heppur.2017.08.049 | | | https://doi.org/10.1016/j.jhazmat.2011.07.114 | | |
| <i>Recovery of cobalt from spent lithium-ion batteries using supercritical carbon dioxide extraction</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>A promising physical method for recovery of LiCoO₂ and graphite from spent lithium-ion batteries: Grinding flotation</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Vacuum pyrolysis and hydrometallurgical process for the recovery of valuable metals from spent lithium-ion batteries</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 589,4 | 2,4 | Total | 195,7 | 0,9 | Total | 414,2 | 3,4 |
| Thermal treatment | 13,8 | 0,8 | Thermal treatment | 0,0 | 0,0 | Thermal treatment | 13,7 | 0,8 |
| Mechanical treatment | 1,1 | 0,1 | Mechanical treatment | 11,7 | 0,7 | Mechanical treatment | 3,0 | 0,2 |
| Chemical treatment | 52,5 | 1,5 | Chemical treatment | 2,6 | 0,2 | Chemical treatment | 56,1 | 2,4 |
| Use of water (washing steps and solutions) | 522,0 | 0,0 | Use of water (washing steps and solutions) | 181,5 | 0,0 | Use of water (washing steps and solutions) | 341,1 | 0,0 |
| 16 | | | 17 | | | 18 | | |
| https://doi.org/10.1016/j.jhazmat.2015.09.050 | | | https://doi.org/10.1016/j.hydromet.2015.09.025 | | | https://doi.org/10.1016/j.jhazmat.2017.05.024 | | |
| <i>Environmentally-friendly oxygen-free roasting/wet magnetic separation technology for in situ recycling cobalt, lithium carbonate and graphite from spent LiCoO₂/graphite lithium batteries</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Thermal treatment process for the recovery of valuable metals from spent lithium-ion batteries</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Recycling metals from lithium ion battery by mechanical separation and vacuum metallurgy</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 144,7 | 7,9 | Total | 183,8 | 3,0 | Total | 30,6 | 1,5 |
| Thermal treatment | 9,1 | 0,5 | Thermal treatment | 14,3 | 0,8 | Thermal treatment | 21,1 | 1,2 |
| Mechanical treatment | 120,7 | 7,1 | Mechanical treatment | 0,7 | 0,0 | Mechanical treatment | 4,5 | 0,3 |
| Chemical treatment | 4,9 | 0,3 | Chemical treatment | 48,7 | 2,1 | Chemical treatment | 0,0 | 0,0 |
| Use of water (washing steps and solutions) | 10,0 | 0,0 | Use of water (washing steps and solutions) | 120,1 | 0,0 | Use of water (washing steps and solutions) | 5,0 | 0,0 |

Table 5 (continued)

| B: PYROMETALLURGICAL METHOD | | | | | | | | |
|---|--------------|----------------------------|--|--------------|----------------------------|---|--------------|----------------------------|
| 1 | | | 2 | | | 3 | | |
| https://doi.org/10.1016/j.susmat.2019.e00139 | | | https://doi.org/10.1016/j.susmat.2019.e00139 | | | https://doi.org/10.1021/acssuschemeng.9b04175 | | |
| <i>Recovery of lithium and cobalt from waste lithium-ion batteries through a selective isolation-suspension approach</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Recovery of lithium and cobalt from waste lithium-ion batteries through a selective isolation-suspension approach</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Alkali Metal Salt Catalyzed Carbothermic Reduction for Sustainable Recovery of LiCoO₂: Accurately Controlled Reduction and Efficient Water Leaching</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 259,3 | 4,2 | Total | 583,1 | 11,3 | Total | 196,8 | 12,2 |
| Thermal treatment | 30,3 | 1,8 | Thermal treatment | 30,3 | 1,8 | Thermal treatment | 144,9 | 8,5 |
| Mechanical treatment | 10,5 | 0,6 | Mechanical treatment | 3,2 | 0,2 | Mechanical treatment | 1,6 | 0,1 |
| Chemical treatment | 34,1 | 1,8 | Chemical treatment | 189,6 | 8,9 | Chemical treatment | 49,3 | 3,6 |
| Use of water (washing steps and solutions) | 184,3 | 0,0 | Use of water (washing steps and solutions) | 352,7 | 0,0 | Use of water (washing steps and solutions) | 1,0 | 0,0 |
| 4 | | | 5 | | | 6 | | |
| https://doi.org/10.1021/acssuschemeng.9b01564 | | | https://doi.org/10.1016/j.minerg.2018.06.023 | | | https://doi.org/10.1016/j.jpowsour.2019.01.072 | | |
| <i>A Simplified Process for Recovery of Li and Co from Spent LiCoO₂ Cathode Using Al Foil As the in Situ Reductant</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Separation of Li and Co from the active mass of spent Li-ion batteries by selective sulfating roasting with sodium bisulfate and water leaching</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Selective extraction of lithium (Li) and preparation of battery grade lithium carbonate (Li₂CO₃) from spent Li-ion batteries in nitrate system</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 576,5 | 14,9 | Total | 97,0 | 5,2 | Total | 742,3 | 11,1 |
| Thermal treatment | 36,4 | 2,1 | Thermal treatment | 52,9 | 3,1 | Thermal treatment | 48,5 | 2,8 |
| Mechanical treatment | 2,0 | 0,1 | Mechanical treatment | 2,2 | 0,1 | Mechanical treatment | 7,5 | 0,4 |

Table 5 (continued)

| | | | | | | | | |
|---|--------------|----------------------------|---|----------------|----------------------------|--|----------------|----------------------------|
| Chemical treatment | 84,4 | 12,6 | Chemical treatment | 41,0 | 2,0 | Chemical treatment | 38,9 | 7,8 |
| Use of water (washing steps and solutions) | 454,4 | 0,0 | Use of water (washing steps and solutions) | 1,0 | 0,0 | Use of water (washing steps and solutions) | 647,4 | 0,0 |
| 7 | | | 8 | | | 9 | | |
| https://doi.org/10.1002/cite.201500066 | | | https://doi.org/10.1016/j.jpowsour.2017.03.093 | | | https://doi.org/10.1016/j.jpowsour.2012.01.152 | | |
| <i>Recovery Concept of Value Metals from Automotive Lithium-Ion Batteries</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>A promising approach for the recovery of high value-added metals from spent lithium-ion batteries</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Development of a recycling process for Li-ion batteries</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 241,1 | 14,2 | Total | 335,5 | 9,2 | Total | 136,3 | 8,7 |
| Thermal treatment | 215,1 | 12,6 | Thermal treatment | 72,5 | 4,3 | Thermal treatment | 105,7 | 6,2 |
| Mechanical treatment | 21,1 | 1,2 | Mechanical treatment | 19,7 | 1,2 | Mechanical treatment | 7,2 | 0,4 |
| Chemical treatment | 4,9 | 0,3 | Chemical treatment | 54,5 | 3,8 | Chemical treatment | 23,4 | 2,1 |
| Use of water (washing steps and solutions) | 0,0 | 0,0 | Use of water (washing steps and solutions) | 188,8 | 0,0 | Use of water (washing steps and solutions) | 0,0 | 0,0 |
| C: DIRECT RECYCLING METHOD | | | | | | | | |
| 1 | | | 2 | | | 3 | | |
| https://doi.org/10.1016/j.joule.2020.10.008 | | | https://doi.org/10.1039/C5GC02650D | | | https://doi.org/10.1039/C6RA27210J | | |
| <i>Efficient Direct Recycling of Lithium-Ion Battery Cathodes by Targeted Healing</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Environmentally friendly recycling and effective repairing of cathode powders from spent LiFePO₄ batteries†</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Direct regeneration of cathode materials from spent lithium iron phosphate batteries using a solid phase sintering method</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 717,3 | 32,5 | Total | 178,871 | 5,706 | Total | 606,692 | 65,672 |
| Thermal treatment | 41,1 | 2,4 | Thermal treatment | 32,0 | 1,9 | Thermal | 220,4 | 12,9 |

(continued on next page)

Table 5 (continued)

| | | | | | | | | |
|---|--------------|---------------|---|----------------|---------------|--|----------------|---------------|
| | | | | | | treatment | | |
| Mechanical treatment | 1,9 | 0,1 | Mechanical treatment | 1,8 | 0,1 | Mechanical treatment | 1,9 | 0,1 |
| Chemical treatment | 670,3 | 30,0 | Chemical treatment | 51,8 | 3,7 | Chemical treatment | 2,4 | 0,1 |
| Use of water (washing steps and solutions) | 4,2 | 0,0 | Use of water (washing steps and solutions) | 93,3 | 0,0 | Use of water (washing steps and solutions) | 382,0 | 0,0 |
| 4 | | | 5 | | | 6 | | |
| https://doi.org/10.1016/j.susmat.2020.e00152 | | | DOI: 10.1039/c7gc02831h | | | DOI: 10.1039/c7gc02831h | | |
| A direct recycling case study from a lithium-ion battery recall | EE (MJ/kg) | CF (kgCO2/kg) | Effective regeneration of LiCoO2 from spent lithium-ion batteries: a direct approach towards high-performance active particles† | EE (MJ/kg) | CF (kgCO2/kg) | Effective regeneration of LiCoO2 from spent lithium-ion batteries: a direct approach towards high-performance active particles † | EE (MJ/kg) | CF (kgCO2/kg) |
| Total | 812,1 | 39,9 | Total | 403,610 | 18,864 | Total | 676,675 | 28,162 |
| Thermal treatment | 164,1 | 9,6 | Thermal treatment | 107,9 | 6,3 | Thermal treatment | 266,2 | 15,6 |
| Mechanical treatment | 270,1 | 15,9 | Mechanical treatment | 2,3 | 0,1 | Mechanical treatment | 2,3 | 0,1 |
| Chemical treatment | 174,7 | 14,4 | Chemical treatment | 290,4 | 12,4 | Chemical treatment | 405,2 | 12,4 |
| Use of water (washing steps and solutions) | 203,3 | 0,0 | Use of water (washing steps and solutions) | 3,1 | 0,0 | Use of water (washing steps and solutions) | 3,1 | 0,0 |

Table 6

Detailed values of embodied energy (EE) and carbon footprint (CF) resulted for all the steps of each considered process (see Ref [1]), divided into four categories: thermal treatments, mechanical treatments, chemicals, and water use. (A) hydrometallurgical processes; (B) pyrometallurgical processes; (C) direct recycling processes. The data were calculated considering the use of distilled water instead of ultrapure water or deionized water for chemicals dilution. In addition, for the products washing, only tap water was considered.

| H: HYDROMETALLURGICAL METHOD | | | | | | | | |
|--|---------------|----------------------------|--|---------------|----------------------------|--|--------------|----------------------------|
| 1 | | | 2 | | | 3 | | |
| https://doi.org/10.1016/j.matchemphys.2017.01.003 | | | DOI: 10.1016/j.jhazmat.2009.11.026 | | | https://doi.org/10.1021/acssuschemeng.7b00571 | | |
| <i>Photocatalytic properties of Co3O4/LiCoO2 recycled from spent lithium-ion batteries using citric acid as leaching agent</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Recovery of cobalt and lithium from spent lithium-ion batteries using organic citric acid as leachant</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Sustainable Recovery of Cathode Materials from Spent Lithium-Ion Batteries Using Lactic Acid Leaching System</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 1602,6 | 69,7 | Total | 1083,4 | 48,4 | Total | 606,7 | 43,4 |
| Thermal treatment | 160,4 | 9,4 | Thermal treatment | 168,0 | 9,9 | Thermal treatment | 165,6 | 9,7 |
| Mechanical treatment | 0 | 0 | Mechanical treatment | 4,1 | 0,2 | Mechanical treatment | 1,2 | 0,1 |
| Chemical treatment | 1441,6 | 60,3 | Chemical treatment | 910,7 | 38,2 | Chemical treatment | 439,3 | 33,5 |
| Use of water (washing steps and solutions) | 0,6 | 0,0 | Use of water (washing steps and solutions) | 0,6 | 0,0 | Use of water (washing steps and solutions) | 0,7 | 0,0 |
| 4 | | | 5 | | | 6 | | |
| https://doi.org/10.1016/j.wasman.2017.03.037 | | | https://doi.org/10.1016/j.jpowsour.2012.06.068 | | | https://doi.org/10.1016/j.cej.2015.06.071 | | |
| <i>Recovery of lithium and cobalt from spent lithium-ion batteries using organic acids: Process optimization and kinetic aspects</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Ascorbic-acid-assisted recovery of cobalt and lithium from spent Li-ion batteries</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Hydrometallurgical processing of spent lithium-ion batteries (LIBs) in the presence of a reducing agent with emphasis on kinetics of leaching</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 1088,8 | 47,7 | Total | 414,0 | 29,1 | Total | 217,1 | 9,4 |
| Thermal treatment | 118,6 | 7,0 | Thermal treatment | 19,8 | 1,2 | Thermal treatment | 97,5 | 5,73 |
| Mechanical treatment | 0,6 | 0,0 | Mechanical treatment | 1,5 | 0,1 | Mechanical treatment | 0,8 | 0,0 |
| Chemical treatment | 969,2 | 40,7 | Chemical treatment | 392,3 | 27,8 | Chemical treatment | 118,2 | 3,56 |
| Use of water (washing steps and solutions) | 0,4 | 0,0 | Use of water (washing steps and solutions) | 0,5 | 0,0 | Use of water (washing steps and solutions) | 0,6 | 0,03 |

(continued on next page)

Table 6 (continued)

| 7 | | | 8 | | | 9 | | |
|--|--------------|----------------------------|--|--------------|----------------------------|---|--------------|----------------------------|
| https://doi.org/10.1016/j.jpowsour.2013.08.128 | | | https://doi.org/10.1016/j.jhazmat.2016.03.062 | | | https://doi.org/10.1016/j.wasman.2016.03.006 | | |
| 128 | | | t.2016.03.062 | | | 2016.03.006 | | |
| <i>Hydrometallurgical process for the recovery of high value metals from spent lithium nickel cobalt aluminium oxide based lithium-ion batteries</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Recycling of spent lithium-ion battery cathode materials by ammoniacal leaching</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>An environmental benign process for cobalt and lithium recovery from spent lithium-ion batteries by mechanochemical approach</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 191,9 | 11,3 | Total | 347,1 | 23,5 | Total | 503,0 | 27,8 |
| Thermal treatment | 155,6 | 9,1 | Thermal treatment | 7,5 | 0,4 | Thermal treatment | 36,5 | 2,1 |
| Mechanical treatment | 1,6 | 0,1 | Mechanical treatment | 1,6 | 0,1 | Mechanical treatment | 75,7 | 4,4 |
| Chemical treatment | 34,4 | 2,0 | Chemical treatment (only the binary ammonia + ammonium sulfite) | 336,7 | 22,9 | Chemical treatment | 390,8 | 21,2 |
| Use of water (washing steps and solutions) | 0,2 | 0,0 | Use of water (washing steps and solutions) | 1,3 | 0,1 | Use of water (washing steps and solutions) | 0,0 | 0,0 |
| 10 | | | 11 | | | 12 | | |
| https://doi.org/10.1016/j.wasman.2017.05.013 | | | https://doi.org/10.1016/j.wasman.2016.09.039 | | | https://doi.org/10.1016/j.jclepro.2015.10.132 | | |
| 13 | | | n.2016.09.039 | | | 15.10.132 | | |
| <i>Recycling of spent lithium-ion battery with polyvinyl chloride by mechanochemical process</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Green and facile method for the recovery of spent Lithium Nickel Manganese Cobalt Oxide (NMC) based Lithium-ion batteries</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>An atom-economic process for the recovery of high value-added metals from spent lithium-ion batteries</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 260,5 | 13,6 | Total | 16,5 | 1,0 | Total | 227,7 | 11,0 |
| Thermal treatment | 42,3 | 2,5 | Thermal treatment | 9,8 | 0,6 | Thermal treatment | 45,6 | 2,7 |
| Mechanical treatment | 147,6 | 8,7 | Mechanical treatment | 1,9 | 0,1 | Mechanical treatment | 0,9 | 0,1 |
| Chemical treatment | 70,4 | 2,4 | Chemical treatment | 4,7 | 0,3 | Chemical treatment | 180,9 | 8,3 |

(continued on next page)

Table 6 (continued)

| | | | | | | | | |
|---|--------------|----------------------------|--|-------------|----------------------------|--|-------------|----------------------------|
| Use of water (washing steps and solutions) | 0,2 | 0,0 | Use of water (washing steps and solutions) | 0,2 | 0,0 | Use of water (washing steps and solutions) | 0,3 | 0,0 |
| 13 | | | 14 | | | 15 | | |
| https://doi.org/10.1016/j.wasman.2016.03.009 | | | https://doi.org/10.1016/j.seppur.2017.08.049 | | | https://doi.org/10.1016/j.jhazmat.2011.07.114 | | |
| Recovery of cobalt from spent lithium-ion batteries using supercritical carbon dioxide extraction | EE (MJ/kg) | CF (kgCO ₂ /kg) | A promising physical method for recovery of LiCoO ₂ and graphite from spent lithium-ion batteries: Grinding flotation | EE (MJ/kg) | CF (kgCO ₂ /kg) | Vacuum pyrolysis and hydrometallurgical process for the recovery of valuable metals from spent lithium-ion batteries | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 67,8 | 2,4 | Total | 14,4 | 0,9 | Total | 73,0 | 3,4 |
| Thermal treatment | 13,8 | 0,8 | Thermal treatment | 0,0 | 0,0 | Thermal treatment | 13,7 | 0,8 |
| Mechanical treatment | 1,1 | 0,1 | Mechanical treatment | 11,7 | 0,7 | Mechanical treatment | 3,0 | 0,2 |
| Chemical treatment | 52,5 | 1,5 | Chemical treatment | 2,6 | 0,2 | Chemical treatment | 56,1 | 2,4 |
| Use of water (washing steps and solutions) | 0,4 | 0,0 | Use of water (washing steps and solutions) | 0,1 | 0,0 | Use of water (washing steps and solutions) | 0,2 | 0,0 |
| 16 | | | 17 | | | 18 | | |
| https://doi.org/10.1016/j.jhazmat.2015.09.050 | | | https://doi.org/10.1016/j.hydro.2015.09.025 | | | https://doi.org/10.1016/j.jhazmat.2017.05.024 | | |
| Environmentally friendly oxygen-free roasting/wet magnetic separation technology for in situ recycling cobalt, lithium carbonate and graphite from spent LiCoO ₂ /graphite lithium batteries | EE (MJ/kg) | CF (kgCO ₂ /kg) | Thermal treatment process for the recovery of valuable metals from spent lithium-ion batteries | EE (MJ/kg) | CF (kgCO ₂ /kg) | Recycling metals from lithium-ion battery by mechanical separation and vacuum metallurgy | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 137,4 | 8,1 | Total | 63,8 | 3,0 | Total | 27,0 | 1,6 |
| Thermal treatment | 9,1 | 0,5 | Thermal treatment | 14,3 | 0,8 | Thermal treatment | 21,1 | 1,2 |
| Mechanical treatment | 120,7 | 7,1 | Mechanical treatment | 0,7 | 0,0 | Mechanical treatment | 4,5 | 0,3 |
| Chemical treatment | 4,9 | 0,3 | Chemical treatment | 48,7 | 2,1 | Chemical treatment | 0,0 | 0,0 |

(continued on next page)

Table 6 (continued)

| | | | | | | | | |
|--|-----|-----|--|-----|-----|--|-----|-----|
| Use of water (washing steps and solutions) | 2,7 | 0,2 | Use of water (washing steps and solutions) | 0,1 | 0,0 | Use of water (washing steps and solutions) | 1,4 | 0,1 |
|--|-----|-----|--|-----|-----|--|-----|-----|

| P: PYROMETALLURGICAL METHOD | | | | | | | | |
|---|--------------|----------------------------|--|--------------|----------------------------|---|--------------|----------------------------|
| 1 | | | 2 | | | 3 | | |
| https://doi.org/10.1016/j.susmat.2019.e00139 | | | https://doi.org/10.1016/j.susmat.2019.e00139 | | | https://doi.org/10.1021/acssuschemeng.9b04175 | | |
| <i>Recovery of lithium and cobalt from waste lithium-ion batteries through a selective isolation-suspension approach</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Recovery of lithium and cobalt from waste lithium-ion batteries through a selective isolation-suspension approach</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Alkali Metal Salt Catalyzed Carbothermic Reduction for Sustainable Recovery of LiCO₂: Accurate) Controlled Reduction and Efficient Water Leaching</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 75,1 | 4,2 | Total | 223,4 | 10,9 | Total | 195,9 | 12,2 |
| Thermal treatment | 30,3 | 1,8 | Thermal treatment | 30,3 | 1,8 | Thermal treatment | 144,9 | 8,5 |
| Mechanical treatment | 10,5 | 0,6 | Mechanical treatment | 3,2 | 0,2 | Mechanical treatment | 1,6 | 0,1 |
| Chemical treatment | 34,1 | 1,8 | Chemical treatment | 189,6 | 8,9 | Chemical treatment | 49,3 | 3,6 |
| Use of water (washing steps and solutions) | 0,2 | 0,0 | Use of water (washing steps and solutions) | 0,3 | 0,0 | Use of water (washing steps and solutions) | 0,1 | 0,0 |
| 4 | | | 5 | | | 6 | | |
| https://doi.org/10.1021/acssuschemeng.9b01564 | | | https://doi.org/10.1016/j.mineneng.2018.06.023 | | | https://doi.org/10.1016/j.jpowsour.2019.01.072 | | |
| <i>A Simplified Process for Recovery of Li and Co from Spent LiCoO₂ Cathode Using Al Foil As the in Situ Reductant</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Separation of Li and Co from the active mass of spent Li-ion batteries by selective sulfating roasting with sodium bisulfate and water leaching</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Selective extraction of lithium (Li) and preparation of battery grade lithium carbonate (Li₂CO₃) from spent Li-ion batteries in nitrate system</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 123,2 | 14,9 | Total | 96,1 | 5,2 | Total | 95,4 | 11,1 |
| Thermal treatment | 36,4 | 2,1 | Thermal treatment | 52,9 | 3,1 | Thermal treatment | 48,5 | 2,8 |
| Mechanical treatment | 2,0 | 0,1 | Mechanical treatment | 2,2 | 0,1 | Mechanical treatment | 7,5 | 0,4 |
| Chemical treatment | 84,4 | 12,6 | Chemical treatment | 41,0 | 2,0 | Chemical treatment | 38,9 | 7,8 |

(continued on next page)

Table 6 (continued)

| | | | | | | | | |
|---|--------------|----------------------------|---|--------------|----------------------------|---|--------------|----------------------------|
| Use of water (washing steps and solutions) | 0,4 | 0,0 | Use of water (washing steps and solutions) | 0,1 | 0,0 | Use of water (washing steps and solutions) | 0,6 | 0,0 |
| 7 | | | 8 | | | 9 | | |
| https://doi.org/10.1002/cite.201500066 | | | https://doi.org/10.1016/j.jpowsour.2017.03.093 | | | https://doi.org/10.1016/j.jpowsour.2012.01.152 | | |
| <i>Recovery Concept of Value Metals from Automotive Lithium-Ion Batteries</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>A promising approach for the recovery of high value-added metals from spent lithium-ion batteries</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Development of a recycling process for Li-ion batteries</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 241,1 | 14,2 | Total | 146,8 | 9,2 | Total | 136,3 | 8,7 |
| Thermal treatment | 215,1 | 12,6 | Thermal treatment | 72,5 | 4,3 | Thermal treatment | 105,7 | 6,2 |
| Mechanical treatment | 21,1 | 1,2 | Mechanical treatment | 19,7 | 1,2 | Mechanical treatment | 7,2 | 0,4 |
| Chemical treatment | 4,9 | 0,3 | Chemical treatment | 54,5 | 3,8 | Chemical treatment | 23,4 | 2,1 |
| Use of water (washing steps and solutions) | 0,0 | 0,0 | Use of water (washing steps and solutions) | 0,2 | 0,0 | Use of water (washing steps and solutions) | 0,0 | 0,0 |

| D: DIRECT RECYCLING METHOD | | | | | | | | |
|---|--------------|----------------------------|---|---------------|----------------------------|--|----------------|----------------------------|
| 1 | | | 2 | | | 3 | | |
| https://doi.org/10.1016/j.joule.2020.10.008 | | | https://doi.org/10.1039/C5GC02650D | | | https://doi.org/10.1039/C6RA27210 | | |
| <i>Efficient Direct Recycling of Lithium-Ion Battery Cathodes by Targeted Healing</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Environmentally friendly recycling and effective repairing of cathode powders from spent LiFePO₄ batteries†</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) | <i>Direct regeneration of cathode materials from spent lithium iron phosphate batteries using a solid phase sintering method</i> | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 713,2 | 32,5 | Total | 85,661 | 5,713 | Total | 224,963 | 13,217 |
| Thermal treatment | 41,1 | 2,4 | Thermal treatment | 32,0 | 1,9 | Thermal treatment | 220,4 | 12,9 |

(continued on next page)

Table 6 (continued)

| | | | | | | | | |
|---|--------------|----------------------------|---|----------------|----------------------------|---|----------------|----------------------------|
| Mechanical treatment | 1,9 | 0,1 | Mechanical treatment | 1,8 | 0,1 | Mechanical treatment | 1,9 | 0,1 |
| Chemical treatment | 670,3 | 30,0 | Chemical treatment | 51,8 | 3,7 | Chemical treatment | 2,4 | 0,1 |
| Use of water (washing steps and solutions) | 0,0 | 0,0 | Use of water (washing steps and solutions) | 0,1 | 0,0 | Use of water (washing steps and solutions) | 0,3 | 0,0 |
| 4 | | | 5 | | | 6 | | |
| https://doi.org/10.1016/j.susmat.2020.e00152 | | | DOI: 10.1039/c7gc02831h | | | DOI: 10.1039/c7gc02831h | | |
| A direct recycling case study from a lithium-ion battery recall | EE (MJ/kg) | CF (kgCO ₂ /kg) | Effective regeneration of LiCoO ₂ from spent lithium-ion batteries: a direct approach towards high-performance active particles† | EE (MJ/kg) | CF (kgCO ₂ /kg) | Effective regeneration of LiCoO ₂ from spent lithium-ion batteries: a direct approach towards high-performance active particles† | EE (MJ/kg) | CF (kgCO ₂ /kg) |
| Total | 609,1 | 39,9 | Total | 400,556 | 18,864 | Total | 673,621 | 28,162 |
| Thermal treatment | 164,1 | 9,6 | Thermal treatment | 107,9 | 6,3 | Thermal treatment | 266,2 | 15,6 |
| Mechanical treatment | 270,1 | 15,9 | Mechanical treatment | 2,3 | 0,1 | Mechanical treatment | 2,3 | 0,1 |
| Chemical treatment | 174,7 | 14,4 | Chemical treatment | 290,4 | 12,4 | Chemical treatment | 405,2 | 12,4 |
| Use of water (washing steps and solutions) | 0,2 | 0,0 | Use of water (washing steps and solutions) | 0,0 | 0,0 | Use of water (washing steps and solutions) | 0,0 | 0,0 |

Fig. 1 reports the relative (A and B) and absolute (C and D) values of embodied energy and carbon footprint, for the 33 considered LIBs recover technologies, evaluated for 1 kg of cathode. The numerical data of absolute values are reported in Table 5.

2. Experimental Design, Materials and Methods

Several recycling technologies have been proposed to recover the valuable materials in spent LIBs. All the processes have been basically classified into three categories: hydrometallurgy, pyrometallurgy and direct recycling [1]. They generally consist of multiple steps, involving chemical, mechanical, and thermal treatments, with also products washing. However, the sustainability evaluation of the global proposed processes is lacking.

ESCAPE method, presented in [1] for the evaluation of materials and/or processes sustainability, considers CO₂ (or carbon) footprint and embodied energy as the only two parameters to be accounted for sustainability analysis. This approach was developed to support design decisions of technologies at low TRL (3–5) or at pilot-scale (TRL 6–8), when a full and exhaustive LCA cannot be realised. In particular, embodied energy of a product refers to all the energies necessary to extract raw materials from minerals and ores, plus the energies used for the final product manufacturing. Carbon footprint corresponds to the greenhouse gases (GHG) generated in material production [4,11].

These parameters, which can be understood by most of the public, were selected on the premise that global warming potential and energy consumption are two of the main LCA impact parameters and that they can be calculated for all life cycle phases of a product/process.

They depend on the selected materials and on the energy for their manufacturing (for example mechanical and/or thermal energy). Since the ESCAPE approach generally refers to laboratory scale processes, electricity is always used to supply energy for both thermal and mechanical treatments. The evaluation of embodied energy and carbon footprint need calculation of the equivalence factors, depending on the fuels input used by countries to produce electricity and the type of energy into which electricity is converted by laboratory instruments [4].

To calculate the equivalence factors, three steps were used:

1. *Calculation of the proportion of electricity obtained from fossil fuels, and nuclear and renewable sources.*

Information on the quantities of electricity produced by the different fuels in the all world countries are available on the reports of the International Energy Agency (IEA) [1,2].

For each country, the proportion of electricity produced from fossil fuels, nuclear power, and renewable sources were calculated by dividing the individual quantities by the total electricity produced. For fossil fuels derived energy, electricity produced from hard coal, brown coal, peat, oil shale and oil sands, coal gases, oil products and natural gas was considered. For renewable sources derived energy, electricity produced from hydroelectric plants, geothermal, solar, wind, tide power and other sources, biofuels, and wastes (including wood waste, other solid waste, and industrial and municipal waste) was considered.

Average energy proportions for all the world have also been calculated, considering the global electricity production. This was considered in the present work.

2. *Calculation of the conversion efficiency of fossil fuels into electricity.*

The second step concerns the calculation of the energy efficiency to generate electricity from fossil fuels, based on the IEA methodology [3]. Data on fuel inputs to public electricity plants and combined heat and power plants, and electricity and heat outputs from these plants were derived from IEA statistics documents. The conversion efficiency of electricity production from fossil fuels can be calculate as [6]:

$$\eta = \frac{E + (H \times \beta)}{F} \quad (1)$$

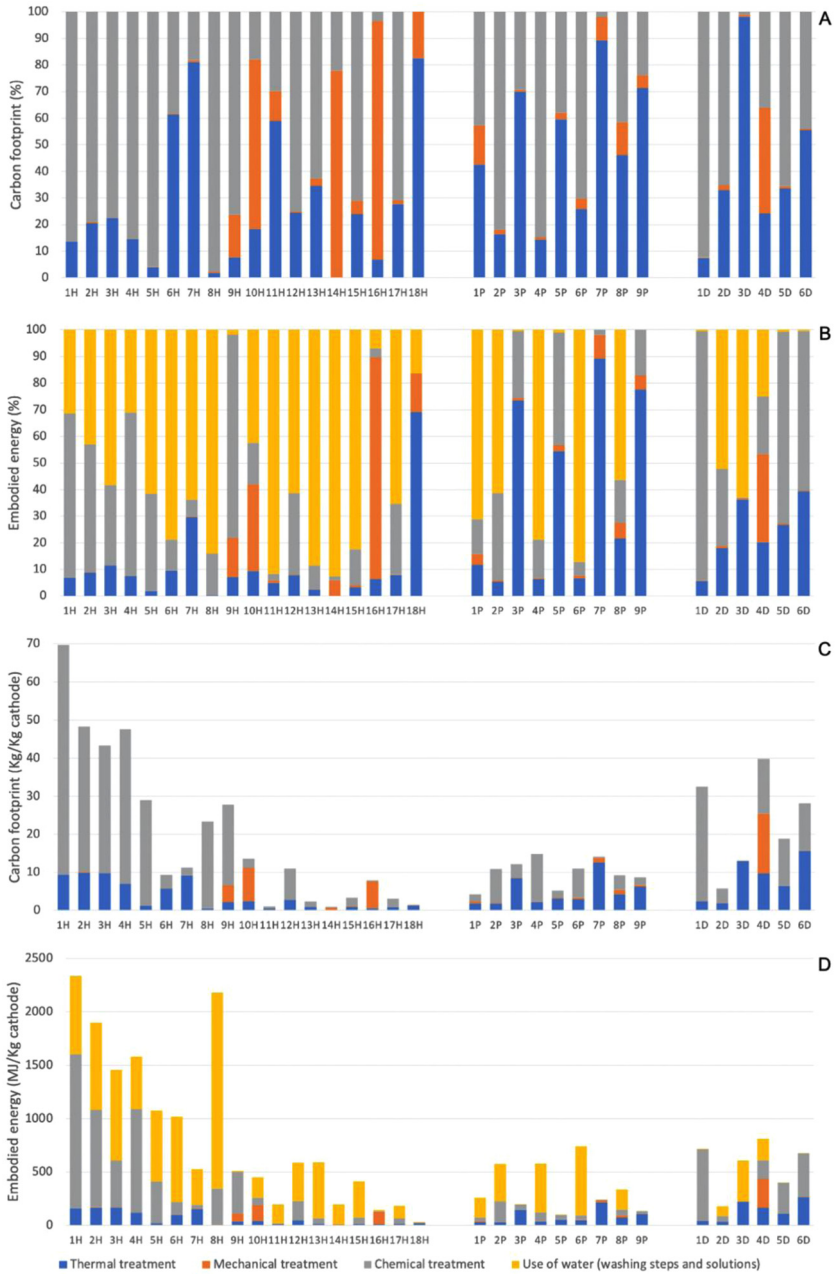


Fig. 1. Relative (A and B) and absolute (C and D) values of EE and CF, for the 33 considered LIBs recover technologies (for the data see Table 5), evaluated for 1 kg of cathode. H stands for hydrometallurgical method; P stands for pyrometallurgical method; D stands for direct recycling.

were,

η = conversion efficiency of electricity production by fossil fuels;

E = electricity production from public electricity plants and public combined heat and power plants;

H = heat output from public combined heat and power plants;

β = loss coefficient. It is expressed as the loss of electricity generation per unit of extracted heat. Its value is assumed to be 0.175 [3];

F = fossil fuel input for public electricity plants and public combined heat and power plants.

3. Calculation of the Country equivalence factors.

Using electricity proportion values between the various fuels and the conversion efficiency calculated previously, for each country the indices of "Energy equivalence (MJ / MJ)" and "CO₂ footprint equivalence (kg / MJ)" were calculated as follows [7]:

$$\text{Energy equivalence} \left(\frac{\text{MJ}}{\text{MJ}} \right) = \frac{\text{Fossil fuel proportion}}{\eta} + \text{Nuclear proportion} + \text{Renewables proportion} \quad (2)$$

$$\text{CO}_2 \text{ footprint equivalence} \left(\frac{\text{Kg}}{\text{MJ}} \right) = \frac{\text{Fossil fuel proportion}}{\eta} \times \text{CO}_2 \text{ conversion factor} \quad (3)$$

where:

CO₂ conversion factor = 0.071 kg/MJ [8]

Once calculated, the equivalence factors are used to evaluate the EE and CF.

To calculate embodied energy and carbon footprint involved in each procedure, it is necessary to know:

- The type of energy generated during the process being studied (like thermal energy in a furnace or mechanical energy in a mixer);
- The instrument operating power;
- The instrument running time (the time of the instrument use).

Then, embodied energy (EE) and carbon footprint (CF) were calculated as follows:

$$EE (J) = \text{Power rating} (W) \times \text{Running time} (s) \times \frac{\text{Energy equivalence}}{\text{Product efficiency}} \quad (4)$$

$$CF (Kg) = \frac{\text{Power rating} (W) \times \text{Running time} (s)}{1 \times 10^6} \times \frac{\text{CO}_2 \text{ footprint equivalence}}{\text{Product efficiency}} \quad (5)$$

The product efficiency depends on the energy that is used:

- For electric to thermal conversion the product efficiency = 1;
- For electric to mechanical (electric motors) conversion the product efficiency = 0.89 [9]

The embodied energy and the carbon footprint (referred to world mean values [2]) calculated for a power rating of 1 W and for a usage of 60 s, considering the global world factors, are shown in Table 4.

In this work the ESCAPE approach is used to evaluate the sustainability of 33 literature processes, proposed for LIBs recovery (see Tables 5 or 6 for the list) [1]. They were selected on the basis of the availability of all the information about the technological steps, in the publication of the methodology sections, to evaluate the embodied energy and the carbon footprint of all the single process. They were calculated using Eqs. (4) and (5). The lists of the parameters used in this work, with corresponding power rating, are reported in Tables 1 and 2.

Along with laboratory procedures, all the reagents employed in the synthesis were considered. The corresponding embodied energy and the carbon footprint are listed in Table 3. Also the water contribution was accounted. On the contrary, for the waste-derived raw materials these

parameters were putted equal to zero, because they origin from other processes as by-products, then it is realistic to neglect emissions and energies associated to their purchase.

The embodied energy and the carbon footprint due to mechanical and/or thermal steps were calculated in accord to the reported procedure. In particular, ESCAPE approach was applied even if several of these technologies were developed only at laboratory scale, at the publication time.

Fig. 1 shows the resulting embodied energy and the carbon footprint evaluated for all the steps of the 33 selected technologies [1], using the ESCAPE approach. The data were calculated considering the processes as exactly described by authors, and they are reported considering separately chemicals, water, mechanical and thermal treatments (see Table 5 for all the data). In particular, in literature, chemicals are often diluted using ultrapure water (dH_2O), that has a high energy impact ($\text{EE}=19.1 \text{ MJ/kg}(\text{dH}_2\text{O})$). Ultrapure water is also sometimes used for washing the final obtained products, contributing to increase the energy impact of the proposed technology. This is extremely evident considering Fig. 1 (data are in Table 5), that shows that the water usage can reach an energy contribution higher than 90% of all process.

It is evident that it is necessary to promote technological improvements able to reduce the environmental impacts, and the ESCAPE approach, allowing to highlight the most onerous steps of a technology, can contribute to provide eco-design strategies. Indeed, the tool allows to rapidly explore alternatives to guide decision-making.

As an example, it is possible to propose the substitution of ultrapure water or deionized water ($\text{EE}=0.24 \text{ MJ/kg}(\text{water})$) with distilled water ($\text{EE}=0.01354 \text{ MJ/kg}(\text{water})$) for chemicals dilution, for all the considered technologies. In addition, for the products washing, only tap water ($\text{EE}=0.005 \text{ MJ/kg}(\text{tap water})$) can be considered. The resulting embodied energy and carbon footprint data are reported in Table 6 (they are also reported and discussed in Fig. 4 of Ref [1]). Comparing Tables 5 and 6, it results evident that the choice to use less onerous water typologies is fundamental to reduce the energy impact of the processes (considering the original technologies, using dH_2O , embodied energy can reach values till $1800 \text{ MJ/Kg}(\text{cathode})$, as in the technology 8H). These results are in accord with literature: LCA data concerning industrial technologies involving chemicals, show similar results, highlighting the high energy involved in ultrapure water usage, and the necessity to replace it with industrial water (tap water) [10]. However, comparing data reported in Table 6, that are obtained by changing only some process steps, with data reported in Table 5, it is possible to highlight that ESCAPE approach allows to rapidly check technological alternatives and support materials selection strategies.

Moreover, being based on only two parameters, the data reported in this paper may be also used by other authors and compared with other approaches developed to evaluate environmental impact. Then it is possible to conclude that ESCAPE tool cannot only be considered as a simple pre-screening methodology, designed for a preliminary sustainability evaluation, but it can also be identified as an eco-design strategy, that can be very useful to guide the decision-making process for a design and/or redesign of a product/technology.

Ethics Statements

This work involves neither human nor animal subjects.

Declaration of Competing Interest

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

Data Availability

Tables 5 and 6 (Original data) (Mendeley Data).

CRediT Author Statement

Serena Ducoli: Investigation, Methodology, Data curation, Writing – review & editing; **Ario Fahimi:** Investigation, Methodology, Writing – original draft; **Elsayed Mousa:** Investigation, Writing – review & editing; **Guozhu Ye:** Writing – review & editing; **Stefania Federici:** Writing – review & editing; **Patrizia Frontera:** Writing – review & editing; **Elza Bontempi:** Conceptualization, Methodology, Writing – original draft, Writing – review & editing, Supervision.

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