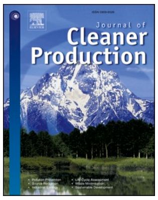
Journal of Cleaner Production 337 (2022) 130454

Contents lists available at ScienceDirect

Journal of Cleaner Production

journal homepage: www.elsevier.com/locate/jclepro



Life cycle assessment of an all-organic battery: Hotspots and opportunities for improvement

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ARTICLE INFO

Handling Editor: Prof. Jiri Jaromir Klemeš

Keywords:
Life cycle assessment (LCA)
Emerging technologies
All-organic battery
Conducting redox polymer based electrodes

ABSTRACT

Organic batteries are emerging as a potential sustainable power source for future flexible devices. Using life cycle assessment, this study analyzed the environmental impacts from the synthesis process for an all-organic battery with conducting redox polymers as active materials for electrodes. Synthesis steps were modeled and analyzed in detail, based on actual laboratory processes data for electrodes, and industrial data for other battery components. Complete and transparent inventory data are presented and can be used in future environmental assessments. The organic battery studied is still at an early development stage, so environmental hotspots and potential improvements in the synthesis processes were examined. For selected environmental impact categories, the life cycle assessment results showed that synthesis of cathode backbone was the major contributor (47–63%) to the environmental impacts of the all-organic battery cell among different synthesis stages, because of a long synthesis route associated with high solvent usage. Solvents (*e.g.*, dichloromethane), catalysts (*e.g.*, copper oxide, Pd(PPh$_3$)$_4$), zinc, and waste treatment processes were important single contributors to the total impacts. The results reveal significant potential for improvement by optimizing the amount of solvents needed to synthesize battery electrodes. Changing treatment methods for laboratory waste solvents can also strongly influence the results.

1. Introduction

Flexible and bendable electronics have undergone rapid development over the past few decades, and are now attracting significant attention from the public and researchers (Delaporte et al., 2020; Lee et al., 2018). In the field of medicine and healthcare, flexible electronics are used extensively as wearable devices to track human activity (*e.g.*, steps, calories burned, sleeping hours) and monitor health conditions (Cima, 2014; Miyamoto et al., 2017; Son et al., 2014). In logistics, intelligent packaging with integrated flexible electronics can provide real-time information on the temperature, humidity, location, and quality of products, allowing optimization of transport, improved product quality, and reduced economic losses (de Abreu et al., 2012; Schaefer and Cheung, 2018; Yam et al., 2005). Flexible and bendable electronics are also expected to be applied in displays such as e-books and e-papers, to meet market and customer requirements (Chung and Kang, 2009). Additionally, application of flexible electronics is contributing to the development of soft robot technologies (Rus and Tolley, 2015). These applications could add much convenience to modern life, enabling a further shift from an individual computing paradigm to a hyper-connected society (Choi, 2014). However, to succeed fully in development of flexible electronics, a more flexible and sustainable power supply unit is needed.

Flexible lithium ion batteries are considered a promising energy storage solution for future flexible devices, as they are thin, lightweight, and have high power output and high energy density (Delaporte et al., 2020; Hu et al., 2010; Nyholm et al., 2011). However, issues relating to the use of relatively scarce lithium and cobalt, immature lithium recycling technology, geopolitical and socio-political problems associated with cobalt mining, and the toxicity of cobalt are major concerns (Janoschka et al., 2012; Larcher and Tarascon, 2015; Muench et al., 2016). Zinc ion batteries, and biodegradable transient batteries are other promising flexible battery technologies that is drawing many researchers' attention (Delaporte et al., 2020; Huang et al., 2019; Zhang et al., 2019). However, human toxicity and other environmental issues (*e.g.*, water, soil, plant pollution) caused by metals used for electrodes and current collector (*e.g.*, Zn, Cu) need to be taken into consideration (Mittal et al., 2021). For example, released sulfate and mobile metals

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https://doi.org/10.1016/j.jclepro.2022.130454
Received 13 July 2021; Received in revised form 7 January 2022; Accepted 7 January 2022

during zinc mining activities can affect human health and the environment directly and indirectly (Mittal et al., 2021; Zhang et al., 2012).

Compared with these battery materials, organic battery materials can be derived from renewable, non-scarce abundant substances extracted from biomass, avoiding mineral resource depletion. Toxicity effects caused by chemicals used in or generated from traditional batteries production are avoided in organic battery, for example, lead, PF_6^-, and HF (Duehnen et al., 2020; Larcher and Tarascon, 2015). End-of-life disposal will also be facilitated by absence of metals, as the battery can be recycled or incinerated using the same waste treatment route as the product it powers. Organic battery materials are also highly versatile, with a tunable structure and bendable characteristics, making them a very promising next-generation flexible energy power source (Song and Zhou, 2013). However, all-organic batteries are less common. Besides, most organic electrode materials do not supply sufficient conductivity, so a large amount of conductive additives and binders are required in the electrodes of organic battery to increase conductivity and sustain material cohesion (Perticarari et al., 2018, 2019; Tong et al., 2019), which complicates the electrode synthesis process and reduced the energy density of the batteries (Emanuelsson et al., 2017). The organic battery cell assessed in this study is based on a newly developed all-organic battery technology using no additives and binders (Strietzel et al., 2020). Hence, it is considered as a promising all-organic battery with low weight, good stability, and fast charging characteristics with potential to be used in flexible devices.

As technology develops, it becomes more costly and difficult to modify material and production process choices when more knowledge and data become available. This means that decisions made at early stages in development of a technology will have far-reaching influences on its environmental performance in technological applications (Arvidsson et al., 2018; Villares et al., 2017). The importance of evaluating the environmental impacts of technologies at an early stage has been widely acknowledged (Hetherington et al., 2014; Sandin et al., 2014). Given the high potential market prospects of all-organic batteries, there is a need to assess the environmental impacts at an early development stage to prevent future unintentional environmental consequences. Life cycle assessment (LCA) is considered a suitable holistic tool for quantifying cumulative environmental loads of products, processes, or activities throughout their life cycle (Hauschild, 2005; Hellweg and Canals, 2014). LCA can cover all stages of a product's life cycle, from raw material extraction to end-of-life disposal or recycling, and can take various environmental impact categories into consideration. This can prevent environmental burden shifting between life cycle stages and environmental impact categories (Kleinekorte et al., 2020). To our knowledge, this is the first LCA study of all-organic batteries. The overall aim of the present study was to provide environmental guidance on future development of an organic battery cell. Specific objectives were (1) to investigate the environmental performance of the all-organic battery described by Strietzel et al. (2020); (2) to identify environmental hotspots in the life cycle stages of this battery; and (3) to identify opportunities to reduce the environmental impacts at an early stage of the development.

2. Materials and method

A cradle-to-factory gate LCA was conducted to quantify the environmental impacts of all synthesis processes of the all-organic battery cell at laboratory-scale. The LCA was conducted in accordance with ISO 14040/44 international standards and the ILCD handbook (Hauschild et al., 2011; ISO, 2006a, 2006b), comprising the four standard phases: goal and scope definition, life cycle inventory (LCI), life cycle impact assessment (LCIA), and interpretation.

2.1. Goal and scope

The goal was to perform a detailed life cycle inventory and assessment of the laboratory-scale synthesis system for an organic battery, to provide environmental guidance in its early-stage development. The functional unit (FU) was one 3 cm by 3 cm organic battery cell prototype. The estimated technology readiness level and manufacturing readiness level are around four. The results were not compared with those of other LCA studies on battery cells, because the organic battery is an emerging technology with unique applications and there is no product with similar applications currently on the market. Hauschild et al. (2018) state that a comparison in LCA is only valid when the products compared provide the same functions. Comparing laboratory-scale processes with industrial-scale processes is another recognized challenge (Hetherington et al., 2014; Pallas et al., 2020; van der Giesen et al., 2020). The environmental impacts calculated from laboratory data are usually much higher than those calculated from industrial data on a mature system (Gavankar et al., 2015). The second functional unit 1 kWh stored electricity is used to facilitate comparison for later lab-scale organic battery studies. Since it is not the aim of the study, the results will not be discussed in this paper, but can be found in Table S3.5. Battery usage and end-of-life treatment stages were not considered in this study. Fig. 1 presents the system boundary of the study, which covered three groups of processes (upstream, laboratory, and downstream):

(1) Upstream processes include raw materials extraction, transportation, battery chemicals manufacturing, and precursor chemicals manufacturing. Raw material extraction refers to acquisition of natural resources for manufacturing chemicals that can be used directly or indirectly in the organic battery cell components. Battery chemicals manufacturing refers to production of chemicals that can be used directly as battery components, *e.g.*, electrolyte, current collector, separator, and battery casing. Precursor chemicals manufacturing refers to production of chemicals needed for synthesizing battery electrodes.
(2) Laboratory processes include synthesis of anode and cathode and assembly of the battery. The laboratory synthesis routes are depicted in Fig. 2.
(3) Downstream processes manage different types of waste flows from upstream processes and laboratory processes.

2.2. Life cycle inventory (LCI) and data sources

The all-organic battery cell assessed in this study was developed by the Nanotechnology and Functional Materials group at Uppsala University (Strietzel et al., 2020), which provided laboratory process data for the present analysis. Data on upstream processes (Section S1 in Supplementary Material) were obtained from the Ecoinvent 3.6 database (cut-off) whenever possible (processes in grey in Fig. 2). Data not available in the database were generated by determining the production routes, based on information from patents and the literature (processes in orange in Fig. 2). Among the data used for downstream processes, parameters for waste solvent distillation processes were taken from Capello et al. (2005) (Table S3.1). Data on waste treatment processes from the Ecoinvent 3.6 database (cut-off) were also used. In the LCI phase of the assessment, the data were complemented with ancillary inputs calculated by parameters from the literature (Capello et al., 2005; Geisler et al., 2004; Piccinno et al., 2016), *e.g.*, energy requirement, cooling water, *etc*. A data inventory for each chemical produced from upstream processes can be found in Tables S1.1-21 in Supplementary Material.

2.2.1. Battery technology and synthesis stages

The battery cell assessed has a mass of 483.23 mg, an active area of 9 cm^2 (3 cm × 3 cm), and a casing area of 12.25 cm^2. The specific capacity is 60 mAh/g of electrode active material, the capacity is 1.08 mAh, the voltage is 0.4 V, and the specific energy is 0.9 Wh/kg. According to Strietzel et al. (2020), the battery can retain 85% of initial capacity after

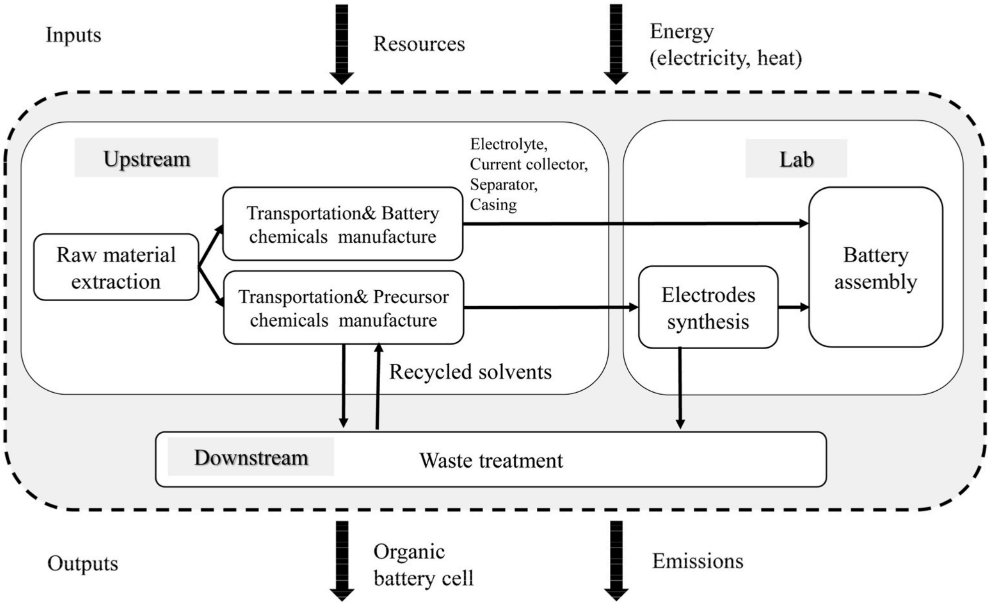

Fig. 1. System boundary (dashed line) of the present study and the three groups of processes covered in the analysis (upstream, laboratory, downstream).

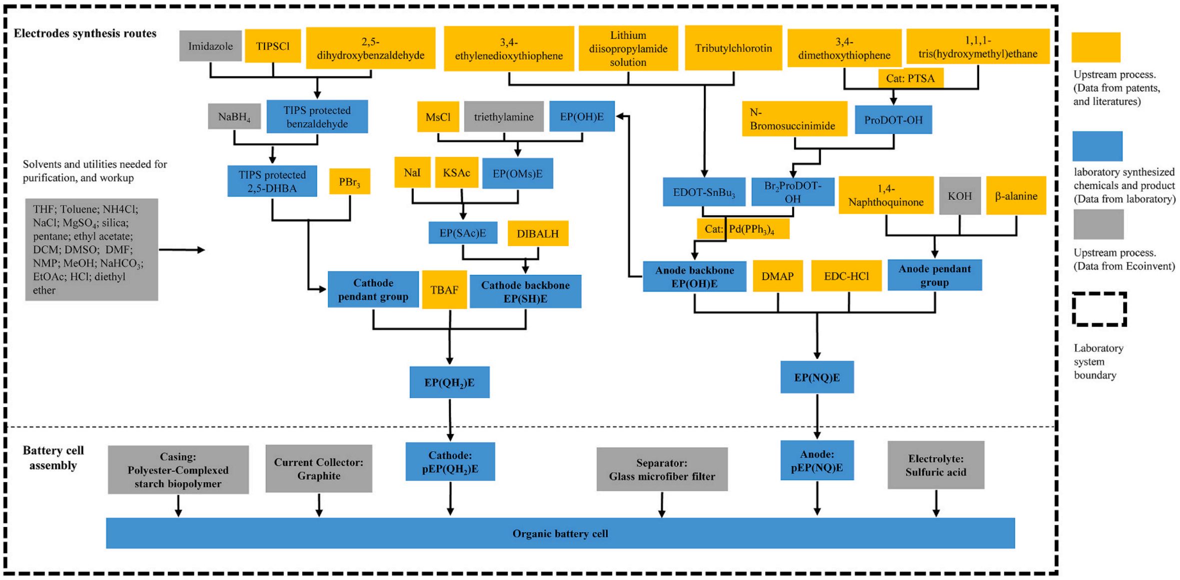

Fig. 2. Laboratory synthesis routes for an organic battery cell. The dashed line divides the laboratory synthesis into electrodes synthesis routes and battery cell assembly. Chemical structures for ProDOT-OH, Br_2ProDOT-OH, EDOT-SnBu3, and TIPS protected 2,5-DHBA can be found in Fig. S2 in Supplementary Material. TIPSCl = Triisopropylsilyl chloride, PTSA = Para-toluenesulfonic acid, MsCl = Methanesulfonyl chloride, KSAc = Potassium thioacetate, DIBALH = Diisobutylaluminum hydride solution, TBAF = Tetra-n-butylammonium fluoride, DMAP = 4-dimethylaminopyridine, EDC-HCl = N-(3-dimethylaminopropyl)-N′-ethylcarbodiimide hydrochloride.

500 charge-discharge cycles, can be fully charged within 100 s, and can maintain good performance at low temperatures, even down to −24 °C.

The battery cell consists of six components: cathode, anode, electrolyte, current collector, separator, and battery cell casing. Table 1 sums up the inventory of one organic battery cell. The electrodes (anode and cathode) consist of two conducting redox polymers (CRPs) as active materials. These two CRPs are composed of polymeric backbones, with trimeric repeat units, and two different pendant groups (naphthoquinone and hydroquinone, respectively). The trimers (EPE) were formed by attaching two 3,4-ethylenedioxythiophene (EDOT, E) at central 3,4-propylenedioxythiophene (ProDOT, P)'s α-positions (Fig. S2, Synthesis of anode backbone). When polymerized polymer backbone serves as to provide electronic conductivity and reduce the solubility of CRPs in the electrolyte. The pendant groups are benzoquinone/hydroquinone (Q/QH_2) and naphthoquinone/naphthohydroquinone (NQ/NQH_2) for cathode and anode, respectively. Pendant groups are capacity carriers in the two electron/two proton ($2e^-/2H^+$) quinone/hydroquinone redox reaction during the organic battery's charge/discharge activity. QH_2 and NQ moieties are attached to the central unity of the trimeric EPE structure, to synthesize the monomers EP(QH_2)E, and EP(NQ)E (Fig. S2). These two monomers are subsequently polymerized to form the CRPs pEP(QH_2)E, and pEP(NQ)E, as the cathode and anode active materials, respectively.

Of the other four battery components, the electrolyte is sulfuric acid (0.5 M), which provides protons, allowing the QH_2 and NQ $2e^-/2H^+$ redox reaction to occur. A glass microfiber filter is used as the separator.

Table 1
Inventory overview for one 3 cm by 3 cm size organic battery cell.

Battery component	Active material	Mass, mg	Inventory process
Cathode	pEP(QH_2)E[a]	18	Own process, based on actual synthesis
Anode	pEP(NQ)E[a]	18	Own process, based on actual synthesis
Current collector	Graphite	9	Market for graphite, battery-grade (global data from Ecoinvent 3.6)
Separator	Glass microfiber filter	46.23	Market for glass fiber (global data from Ecoinvent 3.6)
Electrolyte	0.5 M H_2SO_4	24.5	Market for sulfuric acid (European data from Ecoinvent 3.6)
Battery casing	Biopolymer plastic	367.5	Market for polyester-complexed starch biopolymer (global data from Ecoinvent 3.6)

[a] In pEP(QH_2)E and pEP(NQ)E, E refers to 3,4-ethylenedioxythiophene; NQ refers to naphthoquinone; P refers to 3,4-propylenedioxythiophene; p refers to polymerized; Q refers to benzoquinone; QH_2 refers to hydroquinone.

The current collector is made from Asahi TU-10S carbon conductive paste but, due to lack of data, pure graphite was modeled here as an alternative as Asahi TU-10S carbon paste contains carbon nanoparticles with large amount of graphite particles. The battery cell package is made from plastic (Dupont FEP 500C film), but polyester-complexed starch biopolymer material was used in modeling due to data limitations.

Based on electrode chemical structure (backbone and pendant group), the all-organic battery synthesis routes can be divided into seven synthesis stages: synthesis of anode backbone, synthesis of anode pendant group, synthesis of anode, synthesis of cathode backbone, synthesis of cathode pendant group, synthesis of cathode, and non-electrode components production (Fig. 3). Each synthesis stage includes one or more chemical synthesis processes, as Fig. 3 shows. ProDOT-OH, Br_2ProDOT-OH, EDOT-$SnBu_3$, and EP(OH)E are used in both anode and cathode synthesis. A detailed description of synthesis stages and relevant inputs and outputs flows can be found in Section S2 in Supplementary Material.

2.2.2. *Assumptions*

The following general assumptions were made in the LCI:

1) Swedish and European data from the Ecoinvent 3.6 database were used, when available. Otherwise, global data were used.
2) The reaction yield for chemical products was 95% if process-specific information was missing (Wernet et al., 2012).
3) Electricity at medium voltage from Sweden was used as the electricity source.
4) 100% of the electricity and heat from steam consumed was converted to waste heat, and no heat recovery was assumed.
5) The cooling water was not contaminated during the process, and 100% of the cooling water was returned untreated to the biosphere after use.
6) Metallic catalysts used in industry are likely to be recycled due to their economic value, but catalysts are usually ignored in LCA studies (Parvatker and Eckelman, 2019), which might lead to major uncertainties in the results. Following the suggestion in Piccinno et al. (2016), it was assumed that no catalyst was recycled and a sensitivity analysis was performed to evaluate how this assumption affected the final results.
7) All processes are balanced in terms of inputs and outputs.
8) Waste solvents generated from upstream processes were recycled using distillation, and modeled according to parameters from Capello et al. (2005) (Table S3.1). Waste solvents generated in the laboratory process were treated using the "spent solvent mixture" process from Ecoinvent 3.6 database.

It is worth noting that the above eight assumptions only applied to the processes that we built ourselves. Specifically, assumptions 1, 3, 4, 5, 7 applied to all processes, while assumption 8 only applied to downstream processes and assumptions 2 and 6 only applied to upstream processes.

2.2.3. *Energy requirement*

When data on energy requirement were not available, energy requirement was calculated according to Piccinno et al. (2016) (Equation (1), (3), (4)), and Arvidsson et al. (2014) (Equation (2)). In this study, energy requirement only included the supplied heat (Q_{supply}), and the electricity for stirring (E_{stir}). The supplied heat (Q_{supply}) can be calculated as the sum of energy needed to reach the reaction temperature (Q_{heat}) and the heat loss (Q_{loss}), divided by the efficiency of the heating device (η_{heat}) (Equation (1)).

$$Q_{supply}=\frac{Q_{heat}+Q_{loss}}{\eta_{heat}} \tag{1}$$

The energy needed to reach the reaction temperature Q_{heat} can be calculated from Equation (2).

$$Q_{heat}=C_p{}^*m_{mix}{}^*\Delta T \tag{2}$$

where C_p is specific heat capacity of the main solvent (in J/g*K), m_{mix} is the mass of the reaction mixture (including solvent and reactants), and ΔT is the temperature difference between starting temperature and reaction temperature. If solvent mixtures are used, the specific capacity is estimated using a mole fraction average of each solvent component. Here, it was assumed that reagents were dissolved in solvents for all chemical reactions. To simplify the calculations, the influence of the reagents on the specific capacity of the mixture was neglected.

The energy needed to compensate for the heat loss Q_{loss} is the energy loss from the reactor surface, which can be calculated using equation (3).

$$Q_{loss}=A{}^*\frac{k_a}{s}{}^*\Delta T{}^*t \tag{3}$$

where A is the surface area of the reactor, k_a is the thermal conductivity of the insulation material, s is the thickness of the insulation, ΔT is the temperature difference between the inside and outside of the reactor (ΔT), and t is the reaction temperature. For the reactor-related parameters (A, k_a, s), average data from Piccinno et al. (2016) were used in this study.

Stirring energy can be calculated using equation (4):

$$E_{stir}=\frac{N_p{}^*\rho_{mix}{}^*N^3{}^*d^5{}^*t}{\eta_{stir}} \tag{4}$$

where N_p is power number of the impeller, ρ_{mix} is density of the reaction mixture, N is rotational speed of the agitator, d is diameter of the impeller, t is reaction time, and η_{stir} is efficiency of the agitator. For the impeller-related parameters (N_p, ρ_{mix}, N, d, η_{stir}), average data from Piccinno et al. (2016) were used in this study.

2.3. *Life cycle impact assessment (LCIA)*

The International Reference Life Cycle Data System (ILCD) 2011 midpoint method was used to calculate the environmental impact category indicators. The ILCD method, which was developed by the European Commission - Joint Research Center (EC-JRC), includes 16 recommended impact categories, associated LCIA models, and characterization factors. These recommended LCIA models and characterization factors were selected from existing LCIA methods according to certain criteria for each impact category. (Hauschild et al., 2011). The inventory and the impact calculations were modeled in SimaPro® software.

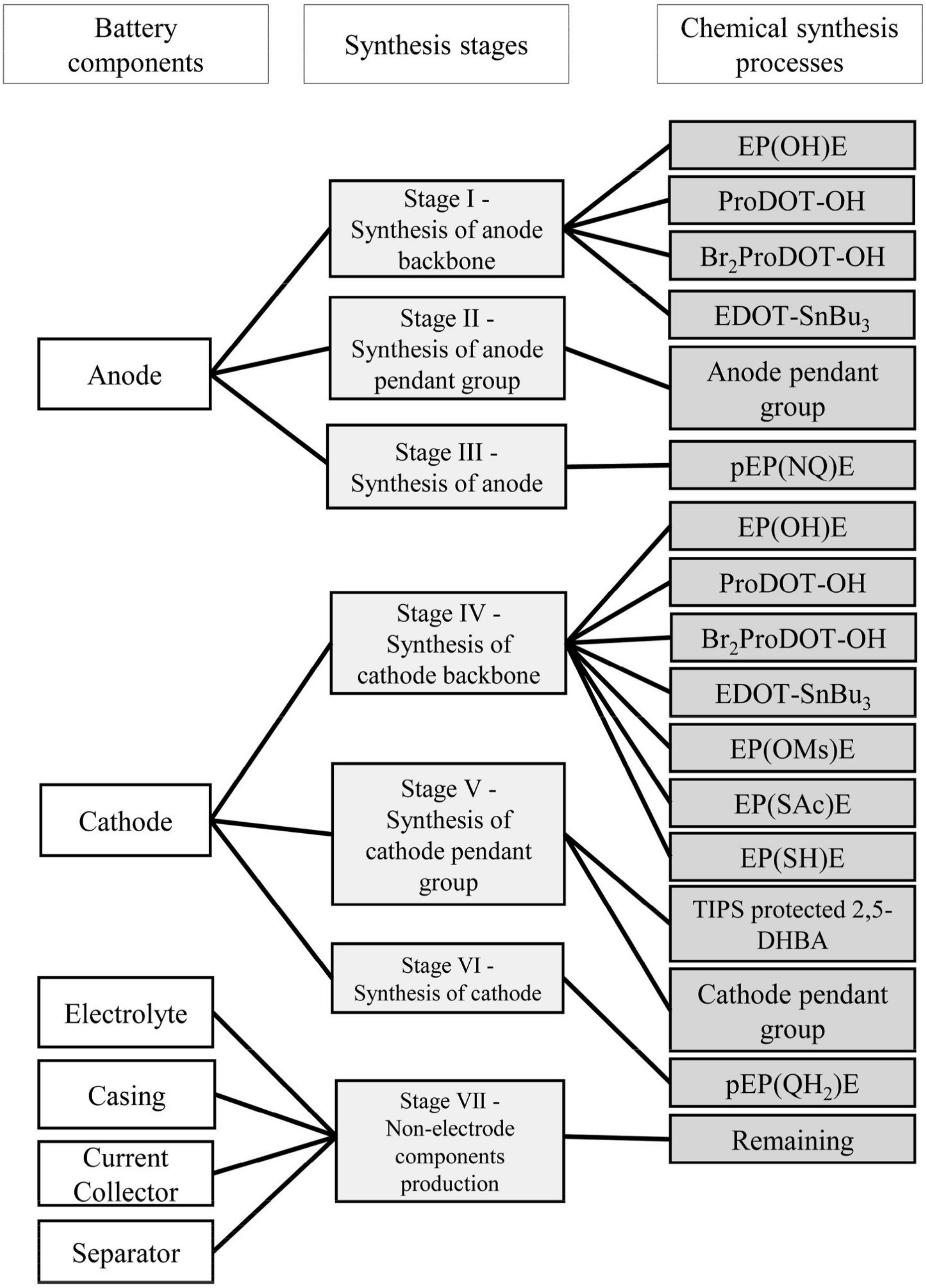

Fig. 3. The chemical synthesis processes are categorized in seven synthesis stages for the assembly of the six components of the all-organic battery cell.

The units of the characterization results vary for each impact category, making it unfeasible to compare them. Normalization and weighting can be applied to determine the most important impact categories for the studied system and for each synthesis stage. The purpose of normalization is to present the relative magnitude of each characterized score by relating it to a common reference system, using normalization factors (Hauschild et al., 2018). The EC-JRC Global normalization factors, which indicate the global impacts per person for each impact category in the reference year 2010 (Benini et al., 2015), were used in this study. For each impact category c, the normalized score (NS_c) was calculated by dividing the impact score (IS_c) by the corresponding normalization factor (NF_c) (Equation (5)):

$$NS_c = IS_c / NF_c \tag{5}$$

In this study, a normalization factor (NF) was calculated for each impact category (c) by dividing the total global impacts of each impact category by the global population (P_g), according to equation (6). The total global impact for each impact category is then the sum of impacts of all elementary flows included in the impact category.

$$NF_c = \frac{\sum_i (CF_i * E_i)}{P_g} \tag{6}$$

where subscript i indicates each elementary flow included in an impact category, CF_i is the characterization factor of elementary flow i, and E_i is the amount of elementary flow i that is released to or extracted from the environment in the reference year 2010 (Hauschild et al., 2018).

Weighting can only be applied based on normalization scores. It supports identification and prioritization of the most relevant impacts categories by applying different weights to each impact indicator (Hauschild et al., 2018). The ILCD weighting factors used in this study include public and LCA expert opinions and the level of impact for each impact category compared to the planetary boundaries (Serenella et al., 2018). Weighted results for each impact category were calculated by multiplying normalized scores of each impact category by the corresponding weighting factors.

The most relevant impact categories for organic battery cell production systems were calculated by following the Product Environmental Footprint Category Rules Guidance (PEFCR), which suggests that the most relevant impact categories should be selected according to the normalized and weighted LCA results, and should be identified as impact categories with a cumulative contribution of more than 80% of the total environmental impacts, without considering toxicity-related impact categories (European Commission, 2018). Following this guidance, four impact categories were considered to be most important in this study: mineral, fossil, and renewable resource depletion (in kg antimony (Sb) eq); climate change (in kg CO_2 eq); ozone depletion (in kg CFC-11 eq); and particulate matter (in kg PM2.5 eq) (Table S3.3 in Supplementary Material).

There is a debate on whether current toxicity-related impact categories are robust enough to be included in LCA (European Commission, 2018; Heijungs et al., 2007; Luca et al., 2016; Vincent-Sweet et al., 2017). This is mainly due to limited elementary flows included in calculation of characterization factors, high uncertainties in modeling fate and exposure, unclear definition of the toxic effects, and unclear mechanisms of the action of metals (Benini et al., 2014; Pizzol et al., 2011). Despite these uncertainties, it is not uncommon for LCA studies to include toxicity-related impact categories in hotspot analysis, to provide more complete results (Gear et al., 2018; Isola et al., 2017; Liu et al., 2015; Troy et al., 2016; Vincent-Sweet et al., 2017). In conventional battery production, metals (*e.g.*, lead (Pb), nickel (Ni), Cu, Al) are usually the main contributors to human or ecological toxicity potential (Ellingsen et al., 2014; Liu et al., 2015; Majeau-Bettez et al., 2011; Wang et al., 2018). No metals are directly used as components in the organic battery cell analyzed in the present study, but some metals are involved in the upstream processes. Therefore, toxicity impacts were included in order to assess whether metals used in the upstream processes, and other chemicals used in the system, contribute significantly to the environmental impacts. Three toxicity-related impact categories were included in the study according to the normalized and weighted results: freshwater ecotoxicity, human toxicity with cancer effects, and human toxicity with non-cancer effects (Table S3.3).

3. Results and discussion

3.1. Midpoint impact results

Results for selected impact categories are summarized in Table 2. All results for ILCD 2011 midpoint categories can be found in Table S3.4-3.5 in Supplementary Material. Overall, electrode (anode and cathodes) synthesis accounted for approximately 99.5–99.9% of the environmental impact for all selected categories, even though electrodes only account for 7% of the total battery cell mass. This is because the inventory data used for electrodes were based on laboratory-scale data, while other battery components were modeled using industrial-scale data. It is worth noting that the mass of electrodes usually account for more than 50% of the total battery cell weight (Deng et al., 2017a; Peters et al., 2016). The low mass proportion of electrodes in the all-organic battery cell is caused by the low technology readiness level of the battery cell and the small scale fabrication technology (laboratory-level). An increase in electrodes' mass percentage can be expected in the future. Previous studies have shown that the environmental impacts of a product decrease with increasing production scale and increasing technological maturity (Gavankar et al., 2015; Troy et al., 2016). Comparing the two different electrodes, the cathode showed higher environmental impacts than the anode for all environmental impact categories, with synthesis stage of cathode backbone being the greatest contributor (Table 2). This was due to the many synthesis steps required in the synthesis stage of cathode backbone, resulting in high consumption of chemicals needed for work-up and purification (*e.g.*, solvents, silicon, Na_2SO_4, etc.) and a larger amount of waste generated.

Fig. 4 summarizes the main contributors for different impact categories. For mineral, fossil, and renewable resource depletion, zinc and catalysts were the greatest contributors. Zinc, a reagent used for producing EDOT (the precursor for the electrode backbone) emerged as the major single contributor, accounting for 52% of the total impact. Production of $Pd(PPh_3)_4$, which is used as a catalyst for synthesizing electrode backbone EP(OH)E, was also responsible for a major share (40%) of the total resource depletion. Therefore, in total more than 90% of the resource depletion was accounted for by anode and cathode backbone synthesis.

For other impact categories, solvent use was clearly the greatest contributor, accounting for 37–100% of the total impact (Fig. 4). Dichloromethane (DCM), ethyl acetate, pentane, tricholoromethane, and tetrahydrofuran (THF) are used as eluents in flash column chromatography for purification and as media for chemical reactions. In general, synthesis stages with purification processes showed higher impact potential, *i.e.*, Synthesis stages of anode backbone, anode, cathode backbone and cathode (Table 2). Synthesis of cathode backbone showed the highest impact of all seven synthesis stages (Table 2), due to the four chemical reactions in cathode backbone synthesis (synthesis process EP(OH)E, ProDOT-OH, EP(OMs)E, and EP(SAc)E) requiring purification processes (Fig. 3). The environmental impacts caused by

Table 2

Life cycle assessment (LCA) characterization results based on one 3 cm by 3 cm organic battery cell as the functional unit (FU), and percentage of the total environmental contribution from different battery components and synthesis stages. * Stage I= Synthesis of anode backbone, Stage II= Synthesis of anode pendant group, Stage III= Synthesis of anode, Stage IV= Synthesis of cathode backbone, Stage V= Synthesis of cathode pendant group, Stage VI= Synthesis of cathode, Stage VII = Non-electrode components production.

Impact category	Value (per FU)	Percentage by synthesis stages						
		Anode			Cathode			Remaining battery components
		Stage I*	Stage II*	Stage III*	Stage IV*	Stage V*	Stage VI*	Stage VII*
Climate change	3.3×10^{-1} kg CO_2 eq	12%	2%	19%	47%	5%	15%	0%
Ozone depletion	1.9×10^{-6} kg CFC-11 eq	7%	3%	1%	62%	4%	22%	0%
Human toxicity, non-cancer effects	6.7×10^{-8} CTUh	19%	2%	15%	51%	4%	9%	0%
Human toxicity, cancer effects	1.3×10^{-8} CTUh	14%	5%	16%	47%	5%	13%	0%
Particulate matter (PM)	2.3×10^{-4} kg PM2.5 eq	9%	2%	13%	52%	5%	19%	0%
Freshwater ecotoxicity	4 CTUe	22%	1%	17%	50%	4%	6%	0%
Mineral, fossil & renewable resource depletion	3.8×10^{-5} kg Sb eq	34%	0%	2%	63%	1%	1%	0%

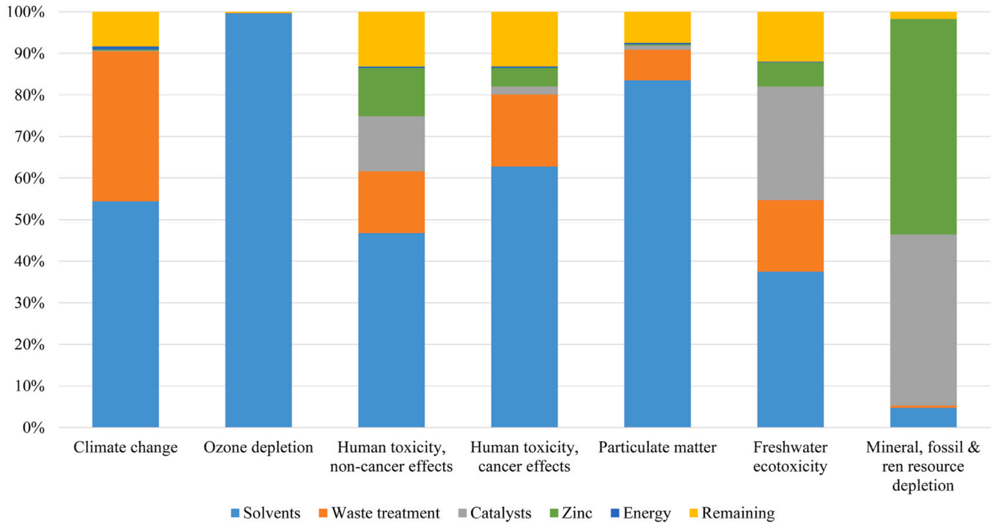

Fig. 4. Contribution of different process flows in synthesis of one 3 cm by 3 cm organic battery cell to selected impact categories. ren. = renewable.

different solvents differ. For example, this study showed that, for the same amount of THF, DCM, and pentane, the climate change caused by THF is the greatest, followed by DCM and pentane. Similar results have been found in a previous study (Amelio et al., 2014).

The ozone depletion category was affected most by use of solvents (Fig. 4). About 99% of the ozone depletion potential was due to use of DCM (82%) and trichloromethane (17%). Dichloromethane is widely used in synthesis stage of cathode backbone for synthesis of intermediate chemical EP(OMs)E, EP(SAc)E, and EP(SH)E, and in synthesis stage of cathode. The increased emissions with use of DCM could affect the rate of stratospheric ozone recovery (Hossaini et al., 2015; Liang et al., 2017). Trichloromethane is used as a solvent in upstream processes, for synthesizing intermediate chemicals ProDOT-OH and EDOT-SnBu$_3$ in anode and cathode backbone synthesis stages, respectively. Pallas et al. (2020) identified trichloromethane as a major contributor to ozone depletion in a solar cell production system. The by-product of trichloromethane production, tetrachloromethane (CCl_4), is also a long-lived ozone-depleting substance (Liang et al., 2017).

The materials used for assembling the all-organic battery cell was clearly lacking toxic materials such as $LiPF_6$, which is used in traditional batteries (Duehnen et al., 2020). However, the solvents and indirectly used metals also contributes to the environmental impact. The result shows that use of solvents was the dominant contributor to freshwater ecotoxicity (37%) and human toxicity with cancer (63%) and non-cancer effects (47%) in the present analysis (Fig. 4). According to Heppel et al. (2011), DCM can be absorbed, distributed, and disseminated rapidly in the human body. Animal tests show that DCM can also cause cancer (Tsai, 2017). Copper oxide (CuO), which is used as a catalyst for producing the precursor (EDOT) of the electrode backbone EP(OH)E, was another major toxicity source, representing 13% and 27% of the impacts for human toxicity with non-cancer effects and freshwater ecotoxicity, respectively. Copper, used as the current collector in other batteries, is often reported to be the main contributor to toxicity-related impact categories (Deng et al., 2017b; Ellingsen et al., 2014; Wang et al., 2020). Again, as it has been discussed before, toxicity related impact categories have higher uncertainty comparing with other environmental impacts.

Waste treatment contributed to climate change (36%), human toxicity with cancer (17%) and non-cancer effects (15%), freshwater ecotoxicity (17%), and particulate matter (7%). There was also a positive correlation between the amount of waste and the amount of solvent used, *i.e.* the amount of waste to be handled increased with increasing solvent use.

3.2. *Energy requirement*

Synthesis of one 3 cm by 3 cm organic battery required 20.2 kJ of heat (generated from steam), and 7.4 kJ of electricity. Heat was used for waste solvent distillation in upstream and downstream processes, and electricity used for cooling or heating chemical reactions, and for waste solvent distillation. Most of the energy requirement was attributable to upstream and downstream processes, including 100% of the heat and 60% of the electricity (Fig. 5). Owing to the relatively low reaction temperatures needed for the chemical reaction steps (usually room temperature), less electricity was required in the laboratory synthesis processes. Due to lack of data, energy requirement was estimated in this study using mathematical equations from small industrial-scale production, which is a source of uncertainty. Therefore, energy requirement in the laboratory synthesis processes might have been underestimated. However, the extremely small overall contribution of energy requirement to different impact categories (Fig. 4) indicates that improving the energy efficiency in future optimization will not contribute substantially to improving the environmental impact.

3.3. *Normalized and weighted results*

The normalized and weighted environmental impacts contributed by each production stage for one 3 cm by 3 cm organic battery are shown in Fig. 6. In most of the synthesis stages, human toxicity with cancer effects was the category making the greatest contribution (21–50% of total impact), except in synthesis stage of anode backbone, and non-electrode components production stage. Freshwater ecotoxicity (10–34%) was another important contributor in most synthesis stages. Resource depletion contributed most in synthesis stage of anode backbone (30%), and was also a dominant contributor to environmental impact in synthesis stage of cathode backbone (19%). Ozone depletion made important contributions in synthesis stage of anode pendant group, cathode backbone, cathode pendant group, and synthesis stage of cathode. In general, climate change, human toxicity with non-cancer effects, and particulate matter had rather low normalized and weighted environmental impacts.

3.4. *Sensitivity analysis*

A number of parameters used in the model have high uncertainty, which means that the parameter value may change and the value used in the model might not always represent reality. To determine how the results were affected by different parameters, a sensitivity analysis was

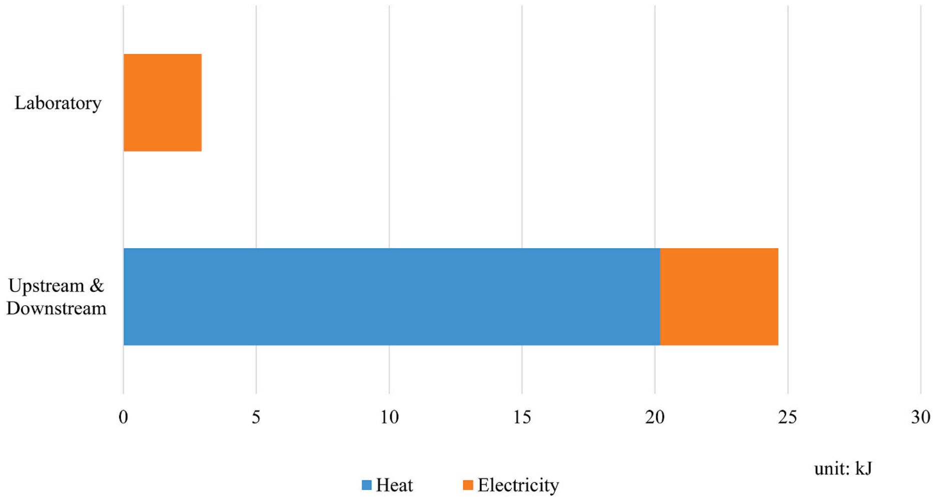

Fig. 5. Energy requirement for synthesizing one 3 cm by 3 cm organic battery cell.

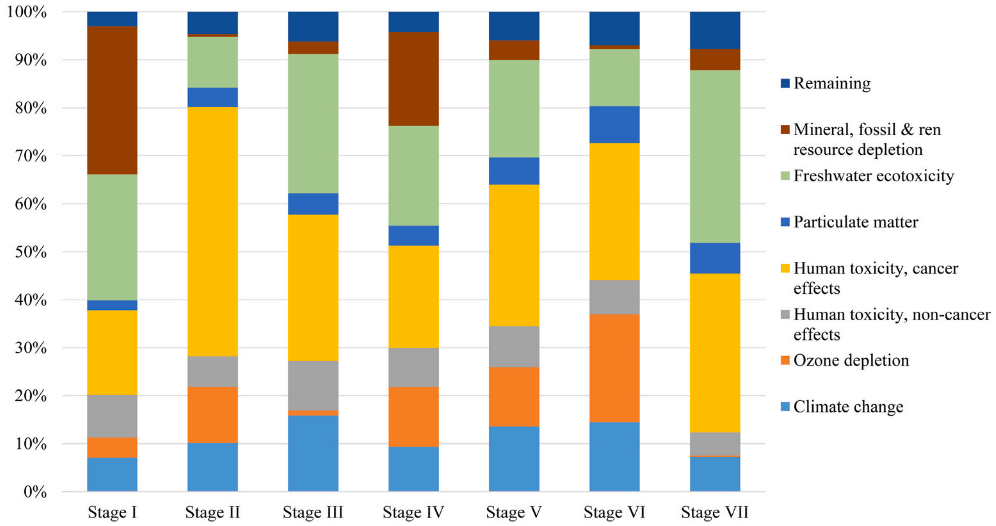

Fig. 6. Normalized and weighted environmental impacts contributed by each production stage (I-VII) in synthesis of one 3 cm by 3 cm organic battery cell. ren. = renewable. * Stage I= Synthesis of anode backbone, Stage II= Synthesis of anode pendant group, Stage III= Synthesis of anode, Stage IV= Synthesis of cathode backbone, Stage V= Synthesis of cathode pendant group, Stage VI= Synthesis of cathode, Stage VII = Non-electrode components production.

performed. The parameters tested in the sensitivity analysis were a) reaction yield, b) catalyst recovery rate (only for catalysts used in upstream processes), and c) laboratory waste solvent treatment method. Furthermore, three scenarios regarding the amount of solvents used at laboratory processes were applied.

It was assumed that the reaction yield for chemical products was 95% when relevant information was missing. Values of 80%, 85%, and 97% were tested in the sensitivity analysis. The results showed that changes in reaction yield had very little effect on the results (<1%) (Table S3.6 in Supplementary Material).

Two catalysts used in the upstream processes are copper oxide and tin(II) chloride ($SnCl_2$). Tests on catalyst recovery rates of 50%, 90%, 99%, and 100% in the sensitivity analysis revealed minor changes (<1%) for all impact categories except freshwater ecotoxicity and human toxicity with non-cancer effects (Table S3.7 in Supplementary Material). The freshwater ecotoxicity impacts decreased by up to 32% and the human toxicity with non-cancer effects decreased by up to 11% when the 100% catalyst recovery rate was applied, due to copper oxide being the main contributor to these two impact categories.

All waste solvents generated in the laboratory processes were assumed to be treated as in the method “spent solvent mixture” from Ecoinvent 3.6 in the baseline scenario. In “Spent solvent mixture” treatment method, around 62% of the solvent were treated with incineration, and rest of the solvent were used as a fuel in cement industries (Valsasina, 2011). Other common technologies to deal with waste solvents include incineration and distillation (Kralisch et al., 2015). Therefore, four waste solvent treatment scenarios were tested in the sensitivity analysis: (1) incineration; (2) recycling by distillation with best-case parameters (Table S3.2 in Supplementary Material); (3) recycling by distillation with average parameters (Table S3.2); and (4) recycling by distillation with worst-case parameters (Table S3.2). The results of the sensitivity analysis are shown in Fig. 7. The results for scenarios (2) and (3) indicated that recycling solvents by distillation can potentially reduce the environmental impacts compared with “spent solvent mixture” treatment and incineration.

In laboratory processes involved in synthesis of the organic battery cell, solvents are used in the synthesis of electrodes’ active materials. Solvent usage in the laboratory is currently not optimized, since the focus of technological development is the performance of the final product. Geisler et al. (2004) estimated the minimum (0.2 kg) and maximum (4 kg) amount of solvent needed for producing one kg of chemical product, according to on-site data from a large size plant and

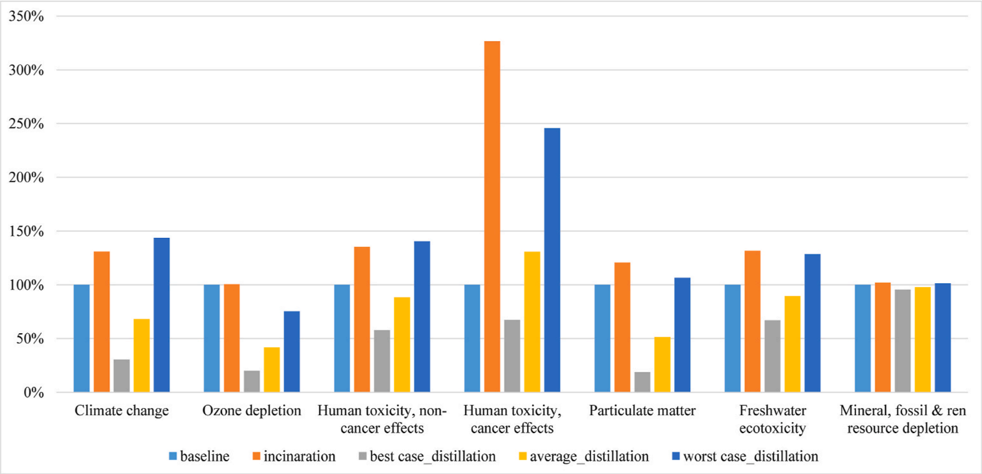

Fig. 7. Influence of different waste solvent treatment methods on the total environmental impact from synthesis of one 3 cm by 3 cm organic battery.

pilot processes, respectively. Piccinno et al. (2016) suggested that a 20% reduction in solvent use is realistic when scaling up from a laboratory process to an industrial process, based on expert opinion. According to the information provided in these two studies, three scenarios were established to assess the influence of excessive solvent use and the potential for improvement:

Scenario 1: 0.2 kg of solvent is used for producing 1 kg of chemical product
Scenario 2: 4 kg of solvent is used for producing 1 kg of chemical product
Scenario 3: A 20% of reduction for laboratory solvent use is applied

The results showed that reduction in environmental impacts under different scenarios vary considerably (Fig. 8). Applying scenario 1 gives the biggest environmental impacts reduction, reducing 5–88% of impacts for all selected impact categories, while applying scenario 3 gives relatively small environmental impacts reduction (Fig. 8). Such difference in results is due to the very high amount of solvents used at laboratory scale, (*e.g.*, usually more than one solvent is used in each chemical synthesis process and single solvent usage can be even up to 80 times of the amount of produced chemical). The rapid reduction of environmental impacts caused by scenario 1 and 2 indicates the big potential in environmental impacts reduction for different environmental impact categories after scaling up the organic battery laboratory system to industrial manufacturing. Additionally, it also indicates the uncertainties of the laboratory-scale LCA, which is in accordance with Pallas et al. (2020), who discussed that improvements on production efficiency should be expected as the emerging technologies scale up from lab to industrial scale.

When scaling up the organic battery laboratory system to industrial manufacturing, improvements in material and energy use efficiency, and the production yield can be expected. The results from three solvent usage scenarios implied that the relative contribution of solvents would most likely be reduced after scaling up to industrial scale production. Consequently, the relative contribution of other process flows will increase and be more visible, like waste treatment processes, energy requirement, nitrogen (used as an inert gas in chemical reactions and solvent recycling processes). Additionally, human toxicity, cancer effects potential, freshwater ecotoxicity potential, and mineral depletion potential can be further reduced if considering catalysts reuse. Even upscaling the laboratory system to industrial production, the electrodes will likely continuously be the dominating contributors among battery components, with the production stage of cathode backbone being the major contributor.

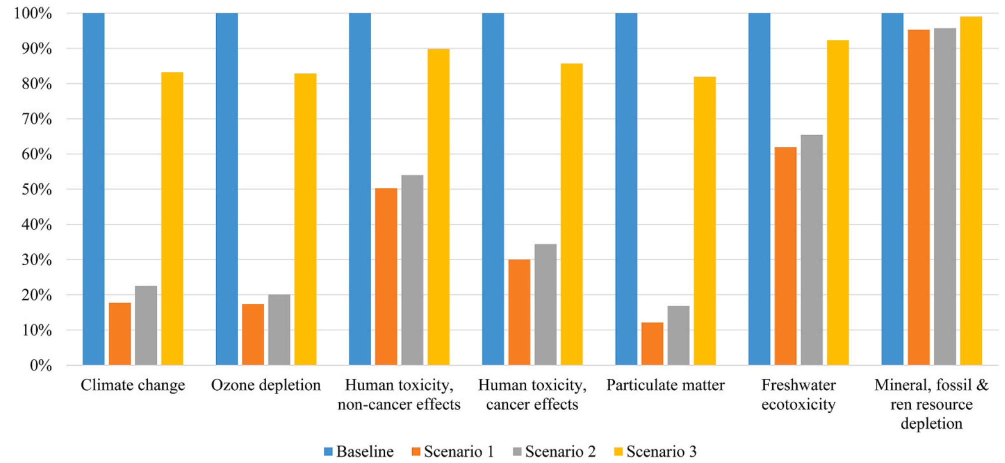

Fig. 8. Influence of amount of solvent use on the environmental impacts from synthesis of one 3 cm by 3 cm organic battery. Scenario 1: 0.2 kg of solvent is used for producing 1 kg of chemical product; Scenario 2: 4 kg of solvent is used for producing 1 kg of chemical product; Scenario 3: A 20% of reduction for laboratory solvent use is applied.

3.5. Opportunities for improvement and future study

Based on the results obtained, solvent use is the hotspot for most environmental impact categories. To reduce the environmental impacts caused by solvent use, the amount of the solvents used in the laboratory needs to be optimized, especially in synthesis stage of anode, cathode and cathode backbone. Recovery of laboratory solvents, *e.g.*, by rotary evaporation, could be another way of reducing environmental impacts, as could using 'green' solvents to replace solvents with high environmental impacts. For example, previous studies recommend use of methanol and acetone to replace DCM, to lower the toxicity (Isola et al., 2017; Montazeri and Eckelman, 2016). According to Cseri. et al. (2018), using solvents like ethyl acetate and toluene is more sustainable than using DCM for the purpose of extraction. Besides, since solvents are used in chemical reactions and purification processes, a shorter synthesis route is an opportunity to reduce the environmental impact from the system, especially for cathode backbone synthesis stage. Production of EDOT (the precursor for the electrode backbone) is also relevant for the overall environmental performance, due to the use of zinc and copper oxide. Reducing the use of EDOT or looking for other alternatives could be a way of reducing the impacts, but would require more detailed research and development. Additionally, looking for other catalysts to replace $Pd(PPh_3)_4$ is a potential way of reducing environmental impacts.

This laboratory-scale LCA provides technology developers with environmental hotspots at the organic battery's early development stage, further guiding the sustainable development of the all-organic battery technology with providing potential opportunities for improvements. Results of this study further contribute to the discussion on the usefulness of lab-scale LCA: laboratory-scale LCA studies should not be used for comparison with established technologies, but to help with the sustainable development of the emerging technologies by identifying the environmental hotspots (Pallas et al., 2020). On the basis of this study, future research on assessing environmental impacts of organic battery at industrial level, with considering the potential improvements of battery performance can be done.

4. Conclusions

This assessment showed that the cathode backbone synthesis stage had the greatest environmental impact (47–63% for different impact categories), due to the long synthesis route and associated large amounts of solvent use and waste. Laboratory solvent use, catalyst use, and waste treatment processes were the main contributing factors to the overall environmental impacts. These results suggest that a shorter synthesis route, optimization of solvent use, recycling of laboratory waste solvents, and reducing or replacing the use of EDOT, and $Pd(PPh_3)_4$ can potentially reduce the overall environmental impacts from synthesis of the organic batteries.

CRediT authorship contribution statement

Shan Zhang: Conceptualization, Methodology, Data curation, Formal analysis, Writing – original draft. **Niclas Ericsson:** Conceptualization, Methodology, Writing - review & editing. **Per-Anders Hansson:** Supervision, Conceptualization. **Martin Sjödin:** Supervision, Investigation. **Åke Nordberg:** Supervision, Conceptualization, Writing - review & editing.

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.

Acknowledgements

This work was supported by Swedish Energy Agency (Project number 48212-1). In addition, we thank Rikard Emanuelsson from Nanotechnology and Functional Materials, Uppsala University, who provided valuable information and data for the study.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jclepro.2022.130454.

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