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New life cycle inventories for end-of-life solid oxide cells based on novel recycling processes for critical solid oxide cell materials

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ABSTRACT

The study presents novel recycling processes for solid oxide cells as well as life cycle assessments (LCA) and life cycle inventory (LCI) analyses. A novel recovery process for the lanthanum-based materials lanthanum oxide and cobalt oxide from the End-of-Life recycling of SOC oxygen electrodes is described and evaluated. In addition, the recovery of yttria-stabilised zirconia and nickel oxide from end-of-life SOC fuel electrodes is presented. Both processes were analysed and a LCI is created for each recycling route. An environmental impact assessment is performed for each recovered material, hotspots and sub-processes are discussed and the results are presented in absolute and relative environmental impact indicator values. The results show that the amount and origin of electricity is one of the most important hotspots in the recycling processes alongside material recovery.

1. Introduction

Hydrogen as an energy vector and hydrogen technologies are increasingly becoming a realistic solution that contributes to the decarbonization of our economy. As fuel cell and hydrogen technologies (FCH) are recognised as one of the key technologies to achieve the EU's sustainable climate targets, more and more of these technologies will be deployed in the near future. FCH technologies will eventually end up in the waste stream and therefore need to be addressed in a sustainable way to ensure circularity through the design of materials and production processes and to minimise the environmental footprint throughout the life cycle. This is not only about ensuring a correct disposal route within a clear legal framework, but also about the complete recovery of critical materials used in FCH technologies [1]. This is crucial not only to ensure that the new FCH technologies and devices have a lower environmental impact throughout their lifecycle than their counterparts, but also to bring highly valuable and rare earths, which are geostrategically important, back into the cycle and thus supporting the entire hydrogen value chain and economy. Key elements for a sustainable End-of-Life (EoL) strategy are: i) inclusion of an assessment of the environmental and socio-economic impact of a recycling technology; ii) a viable supply of recycled materials combined with a recycling infrastructure that uses

economically beneficial and environmentally sound recycling technologies; iii) a clear regulatory strategy for the entire life cycle, including eco-design [2] and producer responsibility extended producer responsibility policy. Even when hydrogen technology eventually comes to market, a strategic approach to the lifecycle of associated devices needs to be built on a solid foundation, which could include the following for the three points above: (i) collection and assessment of data on potential EoL strategies with support for environmentally and economically beneficial recycling technologies demonstrated through Life Cycle Assessment (LCA) methodology for an industry-led roadmap to a regulatory framework for EoL hydrogen appliances. Current and previous EoL experiences with components of similar technical complexity, economic dependency and potential environmental impact (from batteries [3,4] to electronic devices [5], both identified as action areas in the Circular Economy Action Plan 2020) show that waste prevention and management starts with the design of the component and that work also needs to focus on extending the lifetime of devices and ensuring they can be disassembled for reuse, reprocessing and easy recycling. In addition, recycled materials should be used in the manufacture of new appliances without compromising performance and, where possible, at a lower environmental and economic cost. However, this is only possible if this material can be supplied by a reliable supply

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chain that uses environmentally and economically sound recycling technologies.

Solid oxide cells (SOCs) are a group of electrochemical devices within FCH technologies that can be used reversibly as solid oxide fuel cells (SOFCs) to generate electricity and as solid oxide electrolysis cells (SOECs) to produce hydrogen. SOC technology is not yet widely commercialized but is expected to play an important role in the hydrogen economy due to i) the unrivalled conversion efficiency enabled by high operating temperatures [6], ii) the ability to thermally integrate with other processes (e.g. synthetic fuel production) [6], iii) the high flexibility and versatility in component design, manufacturing and component sizing [7,8] and iv) the ability to operate in reversible mode [9]. SOC technology has also improved significantly in recent decades and is approaching the maturity phase [6]. It is therefore expected that the demand for SOC technology will grow rapidly [8], which is also supported by the EU plans for SOC technology research and development targets by Clean Hydrogen Partnership [10]. As the technology has enormous growth potential, it is essential to ensure the environmental and material sustainability of SOCs technologies. The former can be investigated with the help of an environmental life cycle assessment (LCA) and the latter with appropriate EoL management focusing on recycling, circular economy and ecodesign.

The dependence of SOC technology on rare earth metals and other scarce materials could hinder the widespread introduction of this technology. The EU sees dependence on other countries for critical raw materials (CRMs) as a threat to achieving its climate targets [1]. The EU assessment of CRMs for 2023 identified 34 CRMs, some of which are used in SOC technology, namely cobalt, manganese, strontium, lanthanum (LREE) and yttrium (HREE) [1]. In-depth studies on critical materials in hydrogen technologies [11] and in water electrolyzers [12] came to similar conclusions. Lundev et al. [13] investigated critical materials for the energy transition and also identified zirconium, which is used in electrolytes and fuel electrodes of SOCs, as critical. Nickel, which is also an important material for SOCs, is categorized as a strategic material for the EU and not a CRM [1], but is considered critical in other studies [11–13]. Since most SOC components contain some critical materials, SOC research and development is strongly focused on finding substitute materials, reducing the number of critical materials used and increasing recyclability. Therefore, recycling is considered to be one of the most important prerequisites for the introduction of sustainable SOCs technology on a large scale.

Although the awareness of the scientific community and policy-makers regarding the strategic role of CRMs has been increasingly consolidated, in the current literature a very limited number of studies focus on EoL strategies specifically implemented for the recovery and reuse of critical and hazardous materials from EoL SOCs, stated by S. Sarner et al. [14]. S. Saffirio et al. [15] contribute to this field and fill some of the existing gaps, where authors implemented a multi-step recovery strategy of Ni and YSZ from the composite Ni-YSZ phase of EoL SOCs dismantled from stacks after long-time operation by combining hydrothermal treatment and acidic-assisted leaching. The recovery pathway allowed the efficient and selective extraction of Ni from the metal-ceramic composite, to recover a suitable YSZ phase for re-using in novel SOC manufacturing. A similar recovery approach was successfully reported by Yenesew G. et al. [16] for ceramic material of SOC cell components. S. Sarner et al. [14] in recent year have also explored alternative approaches based on the re-oxidation of EoL cells and the recovery of NiO-YSZ composite powders after removing the air electrode by acid leaching and Kaiser C. et al. [17] investigate the removal of the air electrode through ultrasonic de-coating aiming at reducing manual operations and the use of hazardous chemicals. A comprehensive study by Valente et al. [18] examined the consolidated and emerging technologies for the recovery of valuable materials from FCH technologies and concluded that there are currently no developed EoL processes available for SOC components and must take action to extend EoL technologies for SOC recycling.

It is assumed that an environmental impact assessment needs to be performed in all life cycle phases of SOC technologies, with the most suitable methodology being the LCA [19,20], which is highly interconnected in research on FCH technologies and is used to ensure that the technology can support and contribute to the sustainable energy transition. The LCA methodology is recognised as essential in many EU-funded research projects on FCH technologies [21]. A thorough review of existing LCA and sustainability studies on SOC technology was presented by Refs. [22,23]. Mehmeti et al. [22] point out that the lack of up-to-date inventory data is a major problem that affects the accuracy and comparability of existing SOFC LCA studies. The review also found that the disposal and recycling phase is only considered in recent literature and is mostly based on certain assumptions. Longo et al. [24] conducted a cradle to end of life LCA study for SOFCs in residential micro-CHP systems excluding the EoL phase due to the lack of reliable data on future end-of-life processes. The recent study by Ferreira et al. [25] from 2021 shows that the EoL management of SOFC stacks is not well understood due to a lack of information. The authors addressed this issue by modelling different scenarios and assuming a percentage of waste landfilled, incinerated and recycled. A general assumption of 70% recycling was applied only to nickel and YSZ, while LSM and other materials were considered non-recyclable. In many recent LCA studies on SOC technologies (as of 2019), the EoL phase is either not considered [26–29] or insufficient data in the literature is cited as the main reason for omitting the EoL phase [24,30–33]. In the cradle-to-grave studies for SOFC vehicles in Refs. [32,33], the SOFC was not considered when modelling the EoL phase, although other components of the vehicle were included. A comprehensive sustainability assessment of SOC technologies and systems therefore requires adequate modelling of the EoL of SOC, which is currently not available in most of the studies examined.

Table 1 provides an overview of recent LCA studies on SOC technologies, covering the type of study, the scope of the study, the inclusion/exclusion of the EoL phase, the type of the EoL processes used, the EoL modelling approach and the reasons for excluding the EoL phase. It is evident that most of the reviewed articles in which LCA of SOC technology was performed did not consider the EoL phase [24,26–31], or the modelling was severely limited and did not consider the EoL on the SOC level, but rather at the general level of the whole system [32, 33]. The main reason for this was the lack of reliable data, as explained in the studies mentioned above. Also, the studies that considered the SOC-EoL phase only referred to the recycling of the hydrogen electrode and considered the oxygen electrode as non-recyclable [25,34,35].

The main added value of this paper is to present the novel life cycle inventory (LCI) and LCA modelling approaches of recycling processes for solid oxide technology. These processes do not yet exist on a commercial scale and are therefore not used or presented in the reviewed LCA studies on SOC recycling technologies. In order to provide readers (LCA practitioners, recycling industry, researchers in the field of SOC recycling technology, etc.) with more valuable information for further use of the developed LCI, the inventories and data presented in the study are compared with the inventories obtained by reviewing other studies that have conducted LCA of SOC technology. As far as the authors are aware, there is no other complete inventory in the literature for the recycling process of critical raw materials [11]. This study presents novel LCIs, all based on laboratory-level experimental results (TRL3-TRL5), which can be used for LCA studies on SOC technologies for EoL phase application with end-of-life recovery of yttria-stabilised zirconia (YSZ) and nickel oxide (NiO) from EoL SOC fuel electrodes. In addition, a completely novel recovery process for the lanthanum-based materials (lanthanum oxide and cobalt oxide) from SOC oxygen electrode recycling with LCI is presented.

2. Methodology

The methodological approach used in this study to define new LCI for EoL technologies is based on data collection from recycling industry

Table 1
Literature review of recent LCA studies on SOC technologies including the EoL phase with key information on the EoL approach, EoL-LCI, recovery processes and critical materials compared to the present study to emphasize the added value and novelty.

Research	Strazza et al. [34]	Lee et al. [35]	Longo et al. [24]	Bicer & Khalid [26]	Ferreira et al. [25]	Al-Khori et al. [30]	Florio et al. [31]	Vargas & Seabra [33]	Naeini et al. [27]	Zheng et al. [29]	Liao et al. [32]	Jolaoso et al. [28]	This presented study
Year of publishing	2015	2015	2019	2020	2021	2021	2021	2021	2023	2023	2023	2024	2024
Application of SOC	SOFC for distributed power generation	SOFC for CHP power generation	SOFC in residential micro-CHP	SOFC for CHP & power generation - different fuels	SOFC via 3D printing manufacturing operations	SOFC in gas processing operations	SOFC in CHP nano grid system	SOFC in vehicle	SOFC for electricity generation	SOFC in CCHPS	SOFC in vehicle	SOEC for hydrogen production	SOFC
Type of study	Comparative LCA & LCC	LCA	LCA	Comparative LCA	LCA	LCA	Comparative LCA	Comparative LCA	Comparative LCA	LCA + sustainability	Comparative LCA	Comparative LCA	LCA
Scope	Cradle-to-grave	Cradle-to-grave	Cradle-to-end of operation	Cradle-to-end of operation	Cradle-to-grave	Cradle-to-end of operation	Manufacturing + operation	Cradle-to-grave	Cradle-to-product	Manufacturing + operation	Cradle-to-grave	Cradle-to-gate	End-of-life
Inclusion of EoL	yes	yes	no	no	yes	no	no	yes	no	no	yes	no	yes
Inclusion of EoL for SOC	yes	yes	no	no	yes	no	no	no	no	no	no	no	yes
EoL process	Nickel - recycling, Ceramic - landfill	incineration	/	/	Nickel, YSZ - recycling LSM, other - landfill	/	/	/	/	/	/	/	Nickel, YSZ, Lanthanum, Cobalt
EoL Modelling approach	Assumptions (no inventory)	Assumptions	/	/	Assumptions (4 scenarios)	/	/	/	/	/	/	/	Based on laboratory results (TRL3-TRL5
Comments from authors	"After its service life, the SOFC system is considered to be dismantled and the materials recycled or disposed of. It is here assumed that the nickel in the stack will be recycled, whilst for the ceramic stack materials, since a recycling is unlikely, a final disposal as inert materials on a landfill is modelled"	"Recycling (for SOFC) at the end-of-life was not taken into account because of the lack of information."	"The following processes are not included in the analysis: ... - the end-of-life stage: as the examined SOFC system is a prototype and has not yet reached its market maturity, no reliable data regarding future end-of-life processes are available."	"The end-of-life scenarios are not accounted for in this study"	"EoL management of this type of product (SOC) is not well understood due to a lack of information"	"End-of-life was not quantitatively defined because insufficient data were available to quantify the disposal or recycling of SOFC materials properly"	"there is a lack of data about disposal of fuel cell systems" a	"There is absence of information about EoL for the majority of materials in the fuel cell stacks"	"As this is a cradle-to-product analysis, it does not consider the "end of life" of the system"	"The disposal scenarios are not considered in this study"	"Therefore, the disposal phase of the SOFC was not considered in this study" (reason - literature review showed high uncertainty and lack of data)	"The decommissioning of the plant is beyond the scope of this work"	5 newly developed LCI/LCA models for SOC technologies related to EoL phase
Reason for excluding EoL or for relying on assumptions	/	Lack of data	Lack of data	Not given	Lack of data	Lack of data	Lack of data & out of scope	Lack of data	Out of scope	Out of scope	Lack of data	Out of scope	/

experts and chemical experts who have developed recycling processes for SOC technologies at TRL3-TRL5 scale. These laboratory/pilot scale EoL processes were translated into mass and energy balances that serve as input for the definition of the life cycle inventories. Two main recycling routes for exhausted SOC material were investigated, which are shown in Fig. 1: the recycling of lanthanum strontium cobaltite (LSC)-SOC oxygen electrodes with the main output flows of lanthanum oxide (La_2O_3) and cobalt oxide (Co_3O_4) and the recycling of SOC fuel electrodes with an electrolyte consisting of a Ni-YSZ composite with the main output flows of YSZ [36] and NiO. The recycling process for the recovery of YSZ from EoL fuel electrodes at laboratory scale (TRL3) was described by Saffiro et al. [29] and the optimised multistep process formed the basis for scaling up to the pilot-scale plant (TRL5), as described in the public deliverable “Pilot-scale plant (TRL5) based on two integrated existing recycling technologies for SOFCs” [37,38]. In the sub-processes of lanthanum and cobalt recovery via nitric acid, two routes can be distinguished: i) the lanthanum route and ii) the cobalt route (see Fig. 4). The following processes: mechanical detachment, recovery and drying, milling and sieving, acid leaching and centrifugation, and rinsing, recovery and desiccation are part of both recovery

routes, while lanthanum precipitation and calcination to lanthanum oxide (La_2O_3) is only part of the lanthanum route and cobalt precipitation and calcination to cobalt oxide (Co_3O_4) is only part of the cobalt route. The EoL SOCs were provided by Elcogen (Elcogen AS, Tallinn, Estonia) after they were dismantled from the EoL cell stacks and the metal and sealing components were removed. The disassembled EoL cells were then processed to recover the four main materials YSZ, NiO, La_2O_3 and Co_3O_4 as shown in Fig. 1, which also represents the scope of the current study.

2.1. Recycling process of SOC fuel electrode

The multistep recovery pathway is described in detail in by S. Saffiro et al. [39] based on the hydrothermally assisted recovery (HT) process and is shown schematically in Fig. 2. Briefly, after removing the oxygen electrode and the barrier layer, the fuel electrodes and electrolyte are milled and sieved below 25 μm , disaggregated by HT, and then subjected to oxidative acid-leaching to separate YSZ powder and Ni^{2+} ions. The Ni^{2+} containing supernatant was then adjusted to $\text{pH} = 1$, which allowed the precipitation of nickel oxalate ($\text{NiC}_2\text{O}_4 \cdot \text{H}_2\text{O}$) by adding of

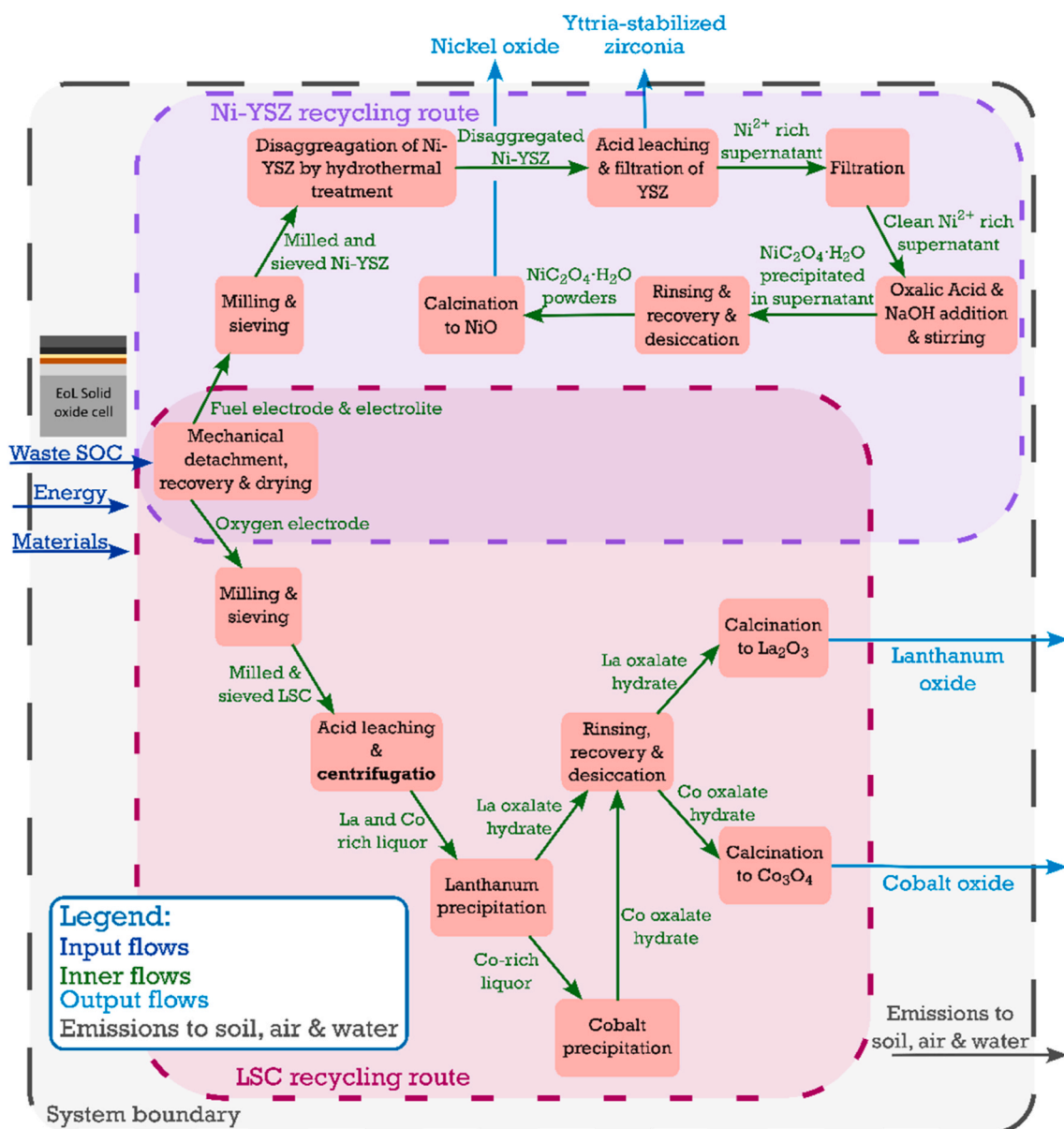


Fig. 1. Boundaries and workflow of the presented LCA study for the recovery of cobalt oxide, lanthanum oxide, NiO and YSZ from EoL solid oxide cells.

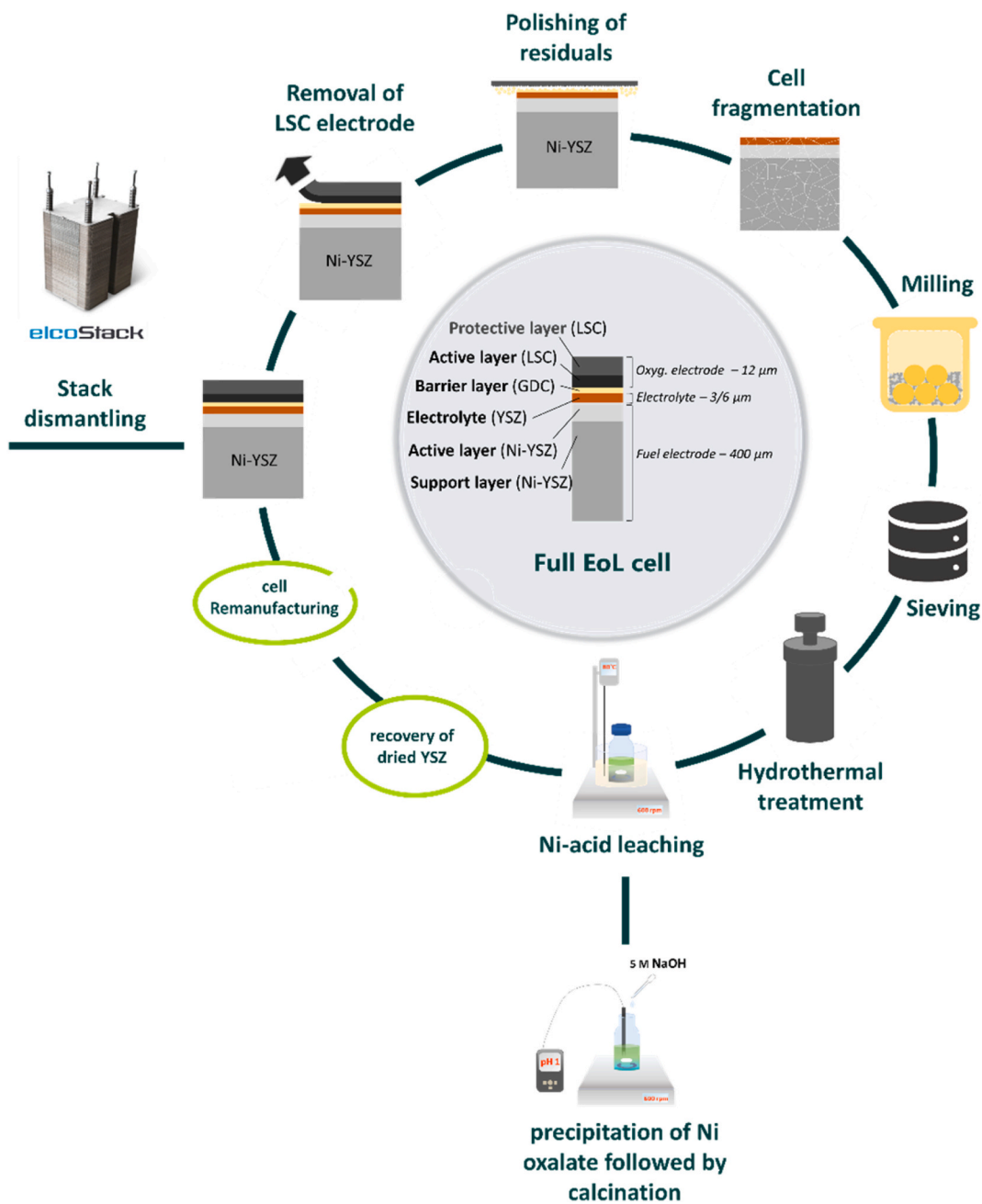


Fig. 2. Schematic representation of the multi-step process for recycling the EoL SOC fuel electrode and electrolyte.

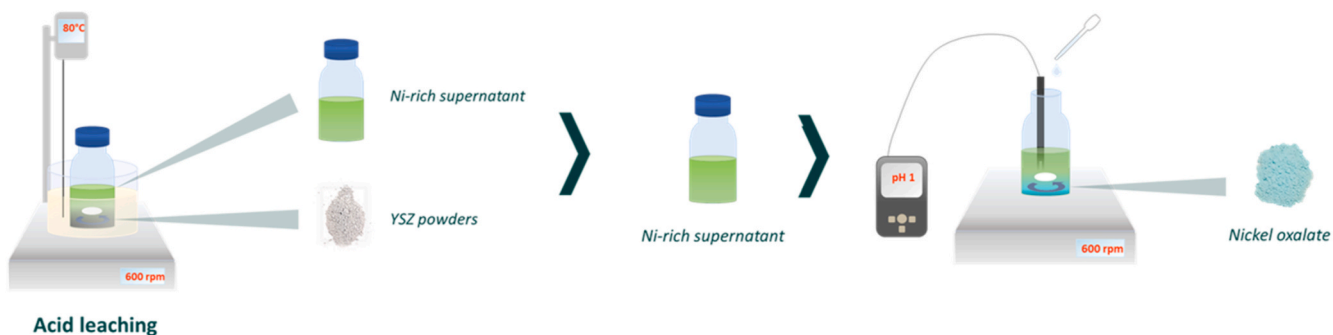


Fig. 3. Schematic of the process for precipitating Ni oxalate from Ni-containing supernatant after acid leaching.

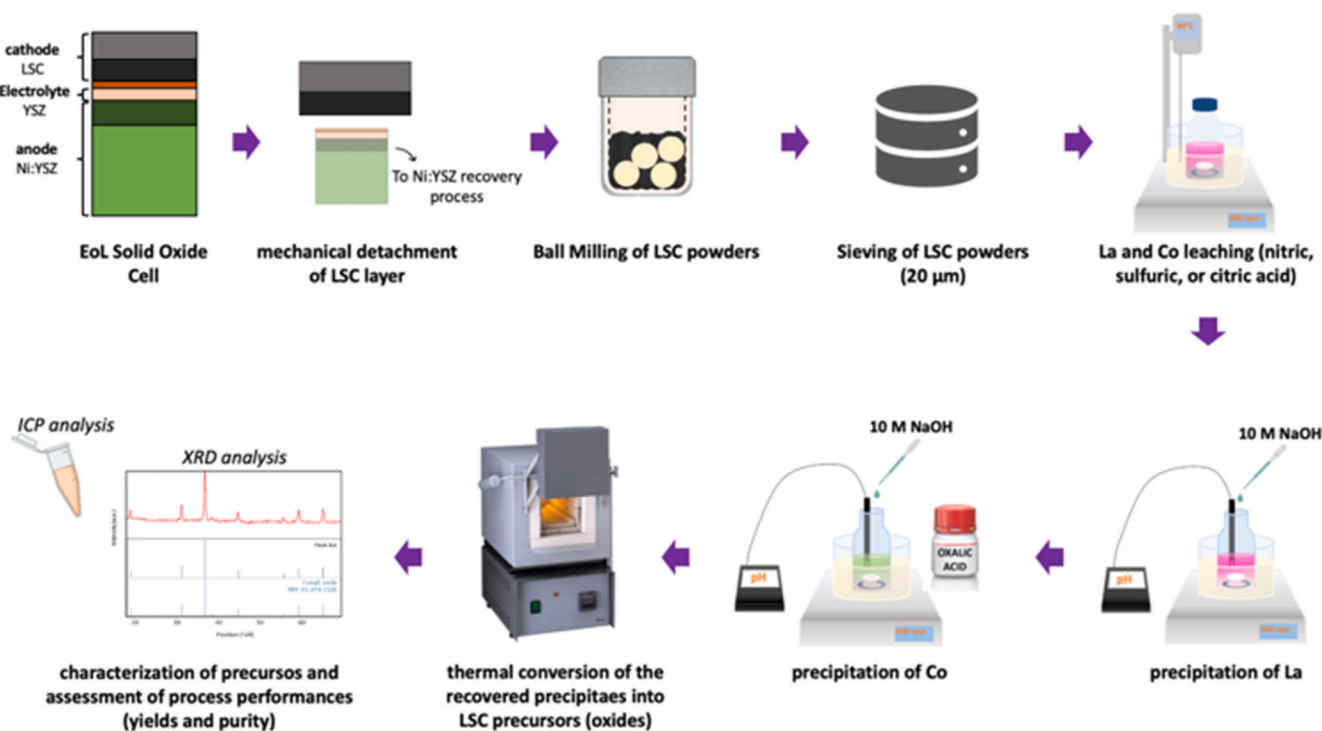


Fig. 4. Workflow for the recovery of lanthanum (La_2O_3) and cobalt (Co_3O_4) from the EoL-SOC cell (TRL3).

oxalic acid, as shown schematically in Fig. 3. The precipitate was then calcined at 450 °C for 3 h to obtain NiO, which serves as a precursor for cell remanufacturing.

The described multistep procedure was carried out on a laboratory scale (TRL3) by processing 2 g of milled and sieved Ni-YSZ powder for each experiment. After optimization and subsequent scale-up to TRL5 [40], 50 g of powder could be processed, combining hydrothermal treatment and acid leaching in a single step.

2.2. Recycling process of SOC oxygen electrode

The provided EoL SOC were first manually scraped to detach the LSC-based electrode and the underlying GDC layer. The LSC is then milled and sieved into dust, which is the main input for nitric acid leaching [36] and the recovery process. Although nitric, sulfuric and citric acids were evaluated in Best4Hy, only the process based on 2 M nitric acid at 80 °C is considered in the environmental assessment as it achieved the best results in terms of recovery and purity of recovered La and Co salts (lanthanum oxide 100% recovery, 96.5% purity; cobalt oxalate 87% recovery, 86% purity). Nitric acid leaching is followed by lanthanum precipitation with oxalic acid and sodium hydroxide, which has two main outputs: cobalt-rich liquor and lanthanum oxalate hydrate. The cobalt-rich lye is participated with oxalic acid and NaOH, yielding cobalt oxalate hydrate. Lanthanum oxalate hydrate and cobalt oxalate hydrate are then rinsed, recovered and desiccated. In the final step, the lanthanum oxalate hydrate is calcined to lanthanum oxide (La_2O_3) and the cobalt oxalate hydrate is calcined to cobalt oxide (Co_3O_4). A more detailed description of the novel EoL SOC oxygen electrode recycling process via the nitric acid route is available in Best4Hy public deliverables D4.2 Technical report on developed recovery technologies for LSC oxygen electrode materials [37] and D4.3 Technical report on lab-scale validation of developed recovery technology for LSC oxygen electrode materials [31,34,35]. Since there are two main outputs (lanthanum oxide, cobalt oxide), cobalt oxide is set as the main output reference flow in the life cycle inventory, which means that all other values of the flows are correlated to obtain 1 g of cobalt oxide. The novel lanthanum strontium cobaltite recycling (LSC – from the oxygen electrode) via the

nitric acid route with the recovery of lanthanum as lanthanum oxide (La_2O_3) and cobalt as cobalt oxide (Co_3O_4) is schematically illustrated at TRL3 level in Fig. 4. The recovery of Sr, Gd and Ce was not targeted in this study or in the Best4Hy project. However, the recovery of Sr was preliminarily investigated; it was performed with low yield (60% recovery, 94% purity) and the recovered oxides were not suitable for conversion into LSC precursors. Therefore, the recovery of Sr, Gd and Ce recovery was not included in the environmental assessment [41,42].

The most important information on the mass balance of the recycling of oxygen electrodes is as follows. The LSC electrode corresponds to a 12 μm thick layer in a SOC. Manual scraping yielded 3.8 g of LSC, which was fed into the acid leaching process. The leached La and Co were recovered by chemical precipitation with oxalic acid (stoichiometric ratio 2:1) and 10 M sodium hydroxide. The nitric acid route yielded 3.2 g lanthanum oxide (100% recovery, 96.5% purity), 2.7 g cobalt oxalate (87% recovery, 86% purity) and 1.21 g strontium oxalate (60% recovery, 94% purity).

3. Life cycle assessment study

The standardized LCA methodology defined in ISO 14040 [41] and ISO 14044 [42] was applied in the study. In addition, the guidelines of the International Reference Life Cycle Data System (ILCD) [43] and the HyGuide, the guide for conducting a life cycle assessment for FCH technologies [44,45], were also taken into account. The LCA methodology described in next chapter consists of four standard phases: (i) goal and scope, (ii) LCI analysis, (iii) life cycle impact assessment (LCIA) and (iv) interpretation of results. All LCA models were created using the LCA tool LCA for experts [46], formerly known as GaBi. The Ecoinvent [47] and Sphera [48] databases were used as background data.

3.1. Goal, scope and boundary conditions

The main objective of this LCA study is to assess the environmental impact of recycling technologies developed for EoL SOC cells, focusing on critical materials within the SOC technology. The environmental LCA study was conducted for the recycling processes of the following critical

SOC materials: (i) lanthanum recovery process from the EoL SOC oxygen electrode as lanthanum oxide (La_2O_3) via nitric acid route at laboratory scale (TRL 3), (ii) cobalt recovery process from the EoL SOC oxygen electrode as cobalt oxide (Co_3O_4) via nitric acid route at laboratory scale (TRL 3), (iii) a process for recovery yttrium-stabilised zirconia (YSZ) and NiO from EoL SOC fuel electrode and electrolyte developed at laboratory scale (TRL 3) and scaled up at pilot scale (TRL 5).

The scope of the environmental LCA study for all four critical material recovery processes is ‘gate-to-gate’, so that only the recovery processes and material flows (from raw materials extraction to the end of material production) in the EoL phase of the SOC EoL cells are assessed, without additional treatment of waste flows. The boundary conditions of the environmental LCA study presented in this paper are divided into two parts: i) SOC fuel electrode and electrolyte material recycling (Ni-YSZ) and ii) SOC oxygen electrode material recycling (LSC).

The functional unit for the recycling of SOC fuel electrode and electrolyte, which also represents the reference flow in the LCA model, is 1 g of recovered NiO and 2.02 g of recovered YSZ at TRL5. The physical and methodological boundaries considered in the LCA of EoL SOC recycling are shown in Table 2.

The functional unit for nitric acid route recycling of the SOC oxygen electrode (LSC), which is also the reference flow in the LCA model, is 1 g of recovered cobalt oxide (Co_3O_4) and 1.36 g of recovered lanthanum oxide (La_2O_3) at TRL3. The physical and methodological boundaries considered in the LCA of recycling of SOC oxygen electrodes for the recovery of cobalt oxide and lanthanum oxide are shown in Table 3.

3.2. Life cycle inventory definition

Life cycle inventories with the most important mass and energy balances are presented and described in detail for all recycling processes. The collection of life cycle inventory data and the definition of LCI is described for two main recycling processes i) recycling of SOC fuel electrodes and ii) recycling of SOC oxygen electrodes, with four main valuable output flows (see Fig. 1).

3.2.1. SOC oxygen electrode recovery inventory

The life cycle inventory of the nitric acid recovery route of lanthanum oxide (La_2O_3) and cobalt oxide (Co_3O_4) is shown in Table 4. Table 4 shows the total values of the aggregated LCI of the chemicals, electricity consumption and databases used for the nitric acid recovery process of the BEST4Hy EoL SOC oxygen electrode (TRL3). The electricity source is a selected average European grid mix for the year 2020 (EU grid mix 2020 [55]). Where possible, the geographical location of the materials used is the European average or a specific country in Europe if data is available. If this was not possible, the global geographical location was chosen (in the case of oxalic acid). The inventory also documents all waste flows that are much larger at laboratory scale than those expected at higher TRL. The processing of waste flows is not considered in the LCA as no data is available due to the low

Table 2

Functional unit, scope and the physical and methodological boundaries considered for the NiO and YSZ recycling process.

NiO and YSZ recycling process	
Functional unit	1 g of recovered NiO and 2.02 g of recovered YSZ
Scope	‘gate to gate’ (EoL phase of SOC cell with YSZ and NiO recovery)
Primary data – foreground system	materials and processes provided within BEST4Hy project [49,50]
Background data – background system	Sphera [51] and Ecoinvent [52]
Life cycle impact assessment	Environmental footprint 3.0 (EF 3.0)
Software used	LCA for experts Sphera software [51]
Recycling efficiencies	90% for YSZ and NiO

Table 3

Functional unit, scope and the physical and methodological boundaries considered in the recycling of SOC oxygen electrodes.

Cobalt oxide and Lanthanum oxide recycling process	
Functional unit	1 g of recovered cobalt oxide (Co_3O_4) and 1.36 g of recovered Lanthanum oxide (La_2O_3)
Scope	‘gate to gate’ (EoL phase of SOC cell with Co_3O_4 and La_2O_3 recovery)
Primary data – foreground system	materials and processes provided within BEST4Hy project [53,54]
Background data – background system	Sphera [51] and Ecoinvent [52]
Life cycle impact assessment	Environmental footprint 3.0 (EF 3.0)
Software used	LCA for experts Sphera software [51]
Recycling efficiencies	Lanthanum: 100%; Cobalt 87%

Table 4

Aggregated LCI for the recovery of lanthanum oxide (La_2O_3) and cobalt oxide (Co_3O_4) from EoL SOC oxygen electrode via the nitric acid route at TRL3.

Material/energy flow	Quantity	Unit	Database used
INPUTS			
EoL SOC Cell	402.56	g	
Deionised H_2O	5298.69	g	RER: Water (deionised) Sphera
Electricity (all steps)	10.453	kWh	RER: Electricity grid mix Sphera
HNO_3 (65%)	43.40	ml	DE: Nitric acid Sphera
H_2O_2 (30%)	9.53	ml	DE: Hydrogen peroxide Sphera
Oxalic acid	6.81	ml	GLO: market for oxalic acid Ecoinvent
NaOH (40%)	59.38	ml	RER: Sodium hydroxide mix Sphera
OUTPUTS			
Lanthanum oxide (valuable)	1.36	g	
Cobalt oxide (valuable)	1	g	
SOC fuel electrode and electrolyte (valuable for NiO-YSZ recycling route)	345.05	g	
sealant removal (waste)	38.34	g	
LSC losses (waste)	1.23	g	
Oversize LSC (>20 μm) (waste) ^a	14.81	g	
Calcination losses (waste)	0.77	g	
Waste solvents and chemicals ^b	119.12	ml	
Deionized water from rinsing	5298.69	g	

^a Oversized LSC powder is not suitable for further treatment at TRL3 level after milling and sieving step. Upscaling will require additional energy to obtain suitable powder for further treatment with larger batches and optimization.

^b Waste solvents and chemicals at TRL3 level are discharged and goes to WWT. Upscaling and optimization for higher TRLs will be carried out as a closed loop.

TRL of the processes.

The LCA model of the novel EoL SOC oxygen electrode recovery with nitric acid route (TRL3) was created using the LCA for experts software and is included in supplementary material (Fig. S1).

3.2.2. SOC fuel electrode and electrolyte recovery inventory

SOC EoL cells are first subjected to detachment of lanthanum strontium cobaltite (LSC64) oxygen electrode (step 1), then the fuel electrode components are milled and sieved below 25 μm (step 2), disaggregated by HT (step 3), and then subjected to acid leaching (step 4) to separate yttria-stabilised zirconia powder and nickel in the form of Ni^{2+} ions in the acid leaching solution. The yttria-stabilised zirconia materials obtained in this route are then recovered (step 5) and Ni is precipitated in the form of precursors (step 6). Then the Ni^{2+} rich supernatant dissolved in HNO_3 is filtered (step 7) and the precipitation process is carried out with oxalic acid and NaOH under stirring (step 8); followed by rinsing, recovery and drying (step 9) to obtain the precipitated

Table 5

Aggregated LCI table for the recovery of 1 g NiO and 2.02 g YSZ from an EoL SOC fuel electrode and electrolyte at TRL5.

Material/energy flow	Quantity	Unit	Used database/description
INPUTS			
EoL SOC cell	5.06	g	
Electricity (all steps)	6.36	kWh	EU-28: Electricity grid mix Sphera
HNO ₃	48.96	g	DE: Nitric acid Sphera
NaOH	9.61	g	EU-28: Sodium hydroxide mix Sphera
Water (deonised)	1568.38	g	EU-28: Water (deionised) Sphera
OUTPUTS			
Recovered NiO (valuable)	1.00	g	
Recovered YSZ (valuable)	2.02	g	
Waste flows			
sealant removal	0.48	g	
cathode detachment (LSC)	0.24	g	
losses after polishing (GDC removal)	0.24	g	
Ni and YSZ losses milling and sieving	0.05	g	
Ni and YSZ HT and recovery losses	0.24	g	
Ni filtration and calcination losses	0.79	g	
Deionized water from rinsing	1568.38	g	
Waste solvents and chemicals ^a	58.57	g	

^a Waste solvents and chemicals at TRL5 level are discharged and goes to WWT. Upscaling and optimization for higher TRLs will be carried out as a closed loop.

NiC₂O₄·H₂O powder. The final step is calcination (step 10) to NiO. Table 5 shows the life cycle inventory for 1 g of recovered NiO and 2.02 g of recovered YSZ for pilot scale (TRL5) EoL processes. The schematic workflow for the existing SOC fuel electrode and electrolyte recovery for YSZ and NiO is shown in Fig. 5 with most important input mass flows and reference output flows - 1 g NiO and 2.02 g YSZ.

The LCA model of EoL SOC oxygen electrode and electrolyte recycling for the recovery of YSZ and NiO using the LCA for experts software is presented in Supplementary materials (Figure S2 and Figure S3).

Based on newly developed life cycle inventories for EoL technologies from SOC, the environmental impact assessment was conducted with a defined boundary condition (EoL phase – Fig. 1) to evaluate the environmental profile of these novel EoL technologies and to identify the main hotspots for further improvement and scaling up of these EoL processes by establishing a first TRL 3 to 5 based LCI for life cycle assessments of the EoL phase.

3.3. Life cycle impact assessment

The Environmental Footprint 3.0 (EF3.0) LCIA method is used in this study to assess the environmental impact of selected recycling technologies. The selection of environmental indicators follows the guidelines of one of the most important documents for LCA of FCH technologies, the HyGuide [56], while the European Commission and the Joint Research Centre (JRC) have supported the EF method quite intensively in recent years. Furthermore, the same LCIA method is also used in the EU project eGHOST [57], which aims to develop new guidelines and improve existing ones (HyGuide) ones for FCH technologies. In this LCA study, we will discuss and analyse the indicators listed in Table 6 used in this study, which were selected based on the literature [58] and the

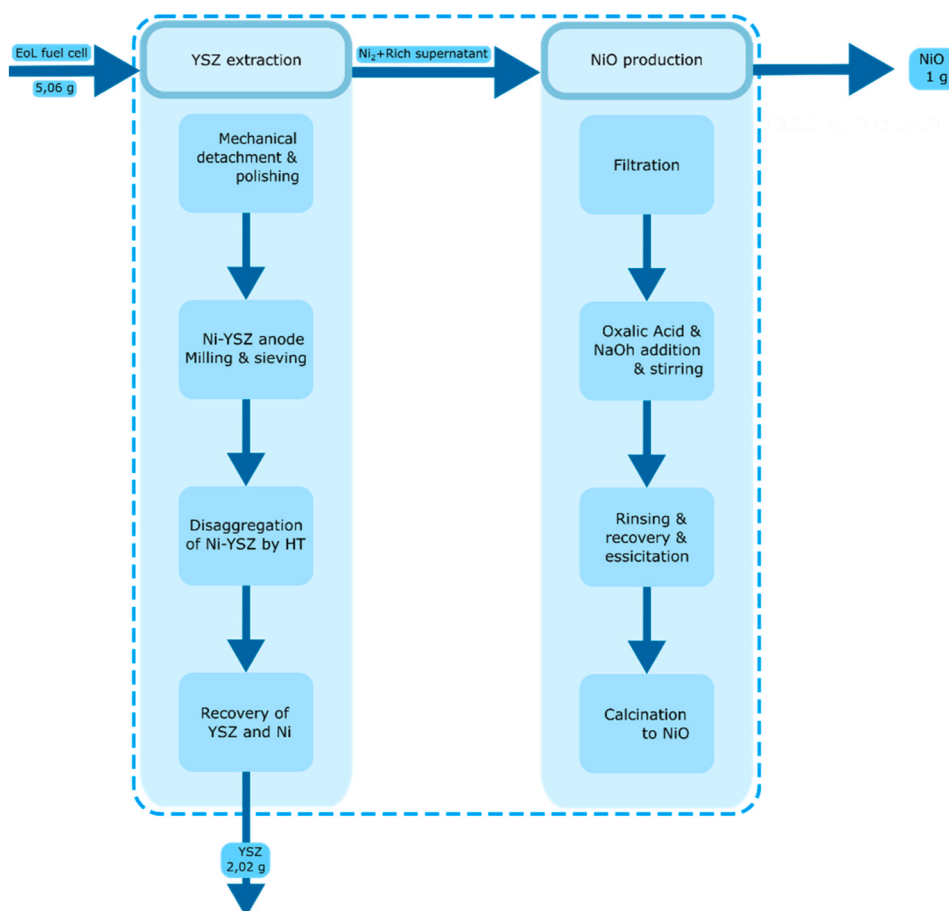


Fig. 5. Schematic workflow for the recovery of YSZ and NiO recovery form EoL SOC fuel electrode and electrolyte with most important mass flows.

Table 6

Environmental impact categories used in this study from EF3.0 LCIA methodology [59].

Impact category	Indicator	Unit
Acidification	Accumulated Exceedance (AE)	mol H + _{eq}
Climate change	Radiative forcing as Global Warming Potential (GWP100)	kg CO₂ eq
Eutrophication (freshwater)	Fraction of nutrients reaching freshwater end compartment (P)	kg P_{eq}
Eutrophication (marine)	Fraction of nutrients reaching marine end compartment (N)	kg N_{eq}
Eutrophication (terrestrial)	Accumulated Exceedance (AE)	mol N_{eq}
Resource use (fossils)	Abiotic resource depletion – fossil fuels (ADP-fossil)	MJ
Resource use (minerals&metals)	Abiotic resource depletion (ADP ultimate reserves)	kg Sb_{eq}

recommendations of the HyGuide, the predecessor of the eGHOST project.

3.4. Interpretation

The interpretation of the results aims to analyse the environmental impacts for all individual recycling processes by identifying hotspots that indicate processes or materials on which FCH and recycling industry need to focus in order to reduce environmental impacts when upscaling to higher TRL. A sensitivity analysis was performed to investigate the influence of the electrical energy source on the environmental impacts. The applicability of newly developed inventories for the recycling of critical materials provides an additional review of the literature and currently available data on the recycling of SOC technologies to illustrate the novelty of the current study. The aim is to provide industry and LCA experts with a guide for future research and improvement of LCI.

4. Results and discussion

This section presents the results of the environmental LCA study with

the identification of the main hotspots within the EoL technology presented above (developed within the BEST4Hy project) for the recovery of SOC critical materials: Lanthanum, Cobalt, Yttrium-stabilised Zirconia (YSZ) and Nickel (Nickel Oxide). The results and the discussion of the environmental impact are presented in the following order.

- (i) In the first part of the results, the environmental impacts of nitric acid recovery route of SOC oxygen electrode material (LSC) from EoL cells for lanthanum as lanthanum oxide (La₂O₃) and cobalt as cobalt oxide (Co₃O₄) at laboratory scale (TRL 3) are presented.
- (ii) In the second part, the environmental impacts of hydrothermally assisted recovery yttrium-stabilised zirconia (YSZ) and nickel as NiO from EoL SOC cell are presented.

As the laboratory work corresponds to TRL 3 and 5, an additional discussion is provided to give the reader useful information on the LCI data. The present work is placed in the context of other work that has been carried out in recent years in the field of LCA of SOC technologies, whereby the added value of the work presented is emphasized in an argumentative manner.

4.1. Environmental impacts from SOC oxygen electrode recycling

The values of the indicators for the environmental impact of the recycling processes for lanthanum oxide and cobalt oxide (see Fig. 4) are given in absolute values in first row (Total) in Table 7, whereby the relative contribution of the sub-processes is also shown in Table 7. In addition, the relative environmental impact of each sub-process is indicated with a hotspot label, with red indicating a high, yellow a medium and green a low environmental impact in relation to the total environmental impact of recycling process. The results are given for the functional unit of the recovered 1.36 g of lanthanum oxide and 1 g of cobalt oxide. Most of the environmental impact within the sub-processes is caused by electricity generation or the extraction and production of raw materials for the chemicals used. The results in Table 7 show that the environmental impacts in all impact categories are more or less evenly distributed across all sub-processes, so that there is no clear hotspot in the sub-processes that would cause the most environmental

Table 7

Total environmental impact of the extraction of 1.36 g lanthanum oxide and 1 g cobalt oxide from the EoL SOC oxygen electrode with additional relative contributions of the sub-processes.

	Acidification [Mole of H+ eq.]	Climate Change – total [kg CO ₂ eq.]	Eutrophication, freshwater [kg P eq.]	Eutrophication, marine [kg N eq.]	Eutrophication, terrestrial [Mole of N eq.]	Resource use, fossils [MJ]	Resource use, mineral and metals [kg Sb eq.]
Total	0.0102 (100%)	3.638 (100%)	3.33E-05 (100%)	2.88E-03 (100%)	0.0308 (100%)	73.5 (100%)	1.23E-06 (100%)
Acid leaching & centrifugation	3%	5%	2%	4%	3%	4%	2%
Calcination to Co₃O₄	10%	13%	5%	8%	8%	14%	6%
Co precipitation	15%	2%	30%	19%	20%	1%	28%
La precipitation	15%	3%	31%	20%	21%	2%	29%
Calcination to La₂O₃	21%	28%	11%	18%	17%	29%	13%
Mechanical detachment, recovery & drying	23%	31%	13%	20%	19%	31%	14%
Milling & sieving	12%	16%	7%	10%	10%	17%	7%
Rinsing, recovery & desiccation	1%	2%	1%	1%	1%	2%	1%

impacts for the entire lanthanum and cobalt recycling process at TRL 3.

Mechanical detachment with recovery and drying contributes the most to the category of impacts on acidification with 23 %, followed by the sub-process of calcination to lanthanum oxide (21 %) and the precipitation steps (15 %). The same two sub-processes also contribute the most to climate change (31 % and 28 % respectively) and resource use – fossils (31% and 29% respectively). Lanthanum precipitation (31 % and 29 % respectively) and cobalt precipitation (30 % and 28 %) are the main causes of eutrophication - freshwater and resource use - minerals and metals. The environmental impacts of eutrophication – marine and eutrophication – terrestrial are most evenly distributed among the sub-processes, with the sub-processes cobalt precipitation, lanthanum precipitation, calcination to lanthanum oxide and calcination to cobalt oxide contributing between 17 % and 21 % of the total impacts. The total climate change of the EoL SOC oxygen electrode lanthanum oxide and cobalt oxide recovery at TRL3 is 3.638 kg CO₂ equivalents for the recovery of 1.36 g lanthanum oxide and 1 g cobalt oxide.

An additional analysis of electricity and chemical consumption for the EoL SOC oxygen electrode recovery process was performed to analyse the contribution of chemical production and electricity consumption to overall environmental impacts (see Fig. 6). The results show that on average 66 % (between 38 % for eutrophication - freshwater and 97 % for resource use, fossils, Fig. 6) of the total environmental impact is due to electricity consumption and 34 % is due to material production and chemicals used in the process. The impacts of resource use – fossil fuels, climate change and acidification are closely linked to electricity consumption, which is why the share of electricity in the above impact categories is 97 %, 95 % and 70 % respectively. In contrast, the impacts in the categories resource use – minerals and metals and eutrophication – freshwater are mainly caused by chemical production, accounting for 58 % and 62 % of the total values respectively.

As electricity consumption is the hotspot in most impact categories, sensitivity analyses were carried out for different electricity mixes (EU grid mix [55] – base case, French grid mix [60] and EU grid mix from renewable energy sources (RES grid mix [61]). The relative environmental impacts of the different electricity mixes used in the recovery process are shown in Fig. 7 whereby the scenario with the greatest environmental impact in the selected impact category was assigned a value of 100%. The results show that climate change is reduced by 86% when using EU electricity mix from renewables compared to using EU

electricity mix. On the other hand, the resource use (minerals and metals) category increase by 127%, as many valuable minerals and metals are used in the production phase of the technologies when using renewable energy sources.

With the future development and higher TRL of lanthanum and cobalt recovery from the EoL SOC oxygen electrode, a lower relative electricity consumption and thus a significant reduction in environmental impact is expected, especially in the categories resource consumption (fossil fuels) and climate change, which are two main hotspots (see Fig. 6) when using EU electricity grid mix.

As illustrated, there is a large energy consumption due to lab-scale research, which represents a critical uncertainty in the LCA. As TRL3-5 does not offer economies of scale, the impact of potential energy reduction from future upscaling will be determined if a larger volume of recycling is expected. 30 %, 50 % and 70 % reduction in energy consumption (EU electricity mix) is defined for three additional scenarios representing upscaling that includes optimization and further reductions. The relative reduction in the impact categories is shown in Table 8, which shows that 70% reduction in electricity consumption in recovery process of lanthanum oxide and cobalt oxide means a reduction in impact categories of –26.5% (eutrophication freshwater) to –67.6% (resource use, fossils).

4.2. Environmental impacts from SOC fuel electrode and electrolyte

The environmental impacts of recycling YSZ and NiO recycling from EoL SOC fuel electrode and electrolyte was assessed for each recycling/recovery process within the EoL phase. The main technological steps and processes have been described in detail in the previous chapters. The main objective here is to assess the environmental impacts of the lab-scale EoL processes and identify the main hotspots, taking into account the limitations of TRL5. Table 9 shows the total environmental impacts for 2.02 g YSZ and 1 g of NiO with relative contributions and hotspots of each EoL sub-process. YZS is recovered in the first step of the process (see Fig. 5), after NiO is recovered in the next step from remaining valuable material input (Ni₂₊ supernatant). ff.

Table 9 shows that in the for the YSZ and NiO-EoL processes, the largest impact is due to the electricity consumption for disaggregation of Ni-YSZ with hydrothermal treatment, which accounts for 62.27% of the total environmental impact on average, followed by the recovery

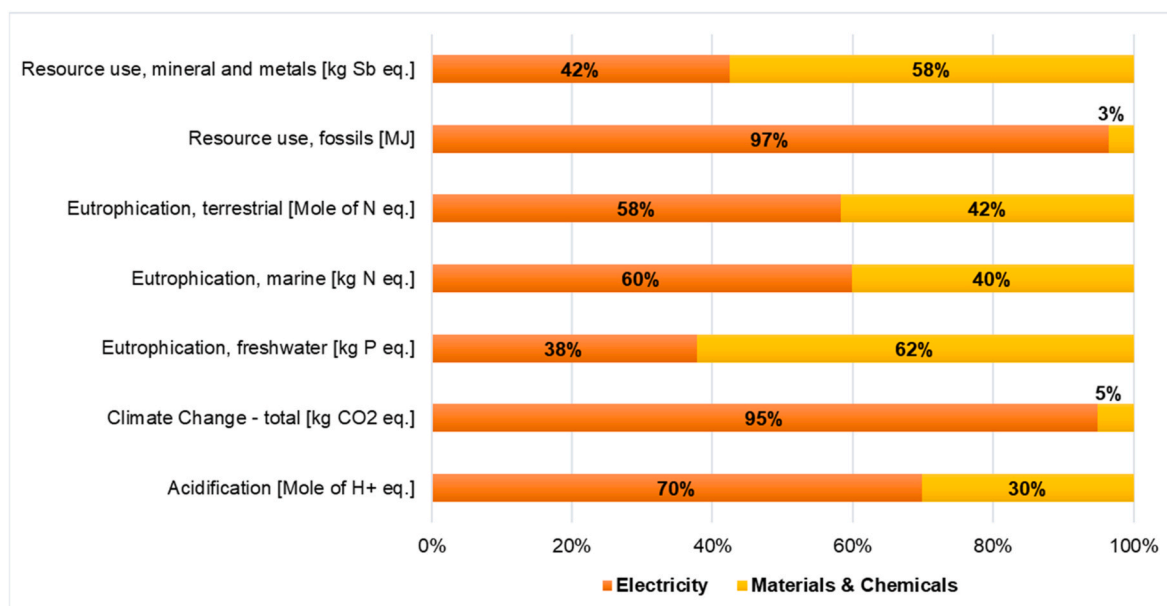


Fig. 6. Contribution to the total environmental impact due to electricity consumption (EU grid mix) and materials/chemicals used in the recovery of lanthanum oxide and cobalt oxide.

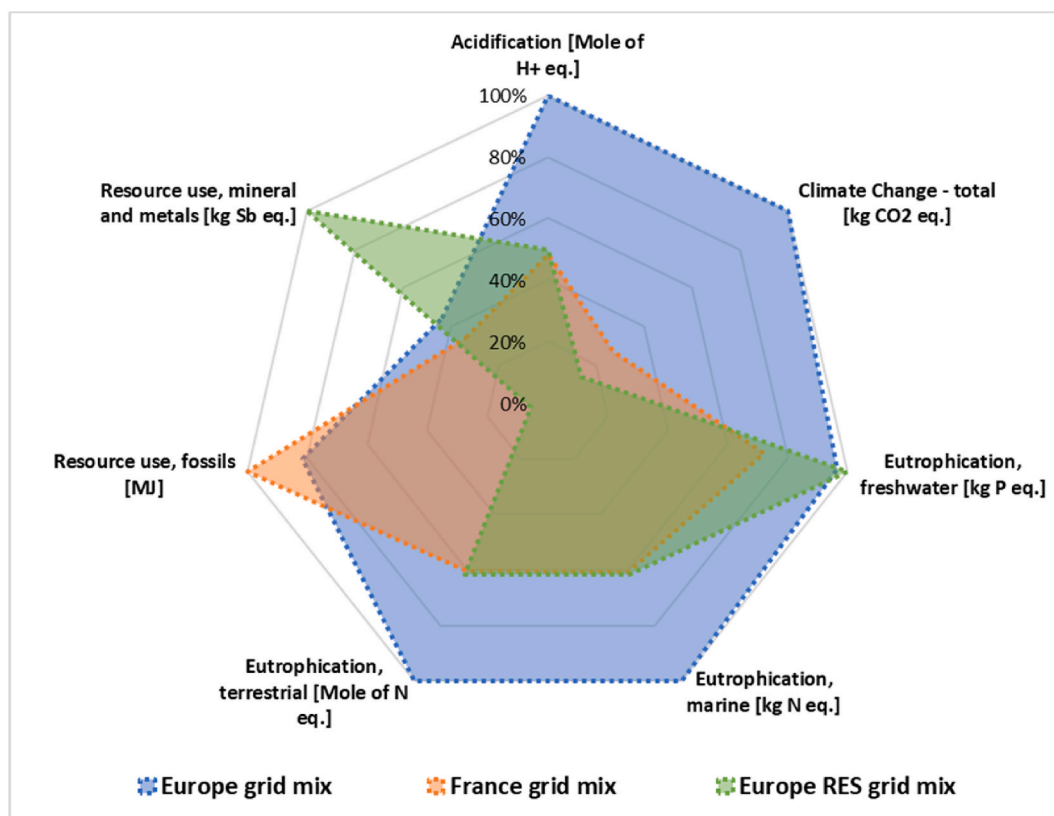


Fig. 7. Relative environmental impacts of recycling 1.36 g of lanthanum oxide and 1 g of cobalt oxide recycling processes with EU electricity, French electricity and RES EU electricity mix.

Table 8

Reduction of environmental impacts per FU with energy consumption reduction due to economy of scale.

	Total per FU 1.36 g La ₂ O ₃ and 1 g Co ₃ O ₄	30% El. reduction	50% El. reduction	70% El. reduction
Acidification [Mole of H+ eq.]	100.0%	-21.0%	-35.0%	-49.0%
Climate Change - total [kg CO ₂ eq.]	100.0%	-28.4%	-47.4%	-66.4%
Eutrophication, freshwater [kg P eq.]	100.0%	-11.3%	-18.9%	-26.5%
Eutrophication, marine [kg N eq.]	100.0%	-18.0%	-29.9%	-41.9%
Eutrophication, terrestrial [Mole of N eq.]	100.0%	-17.5%	-29.2%	-40.8%
Resource use, fossils [MJ]	100.0%	-29.0%	-48.3%	-67.6%
Resource use, mineral and metals [kg Sb eq.]	100.0%	-12.7%	-21.2%	-29.7%

process with 19.87%, which are both processes in the first step of the recycling process that recovers YSZ. The process “NaOH addition with stirring” accounts for 15.47% of the total environmental impacts on average, but it is the process in step 2 that follows the process of YSZ recycling. All other processes for YSZ and NiO recycling account for less than 1.4% of the total environmental impacts on average: milling and sieving – 1.37%, mechanical detachment – 0.63%, rinsing, recovery & drying – 0.29% and calcination – 0.1%. If we compare the shares of the total environmental impacts of the 1st step (recovery of YSZ) and 2nd step (recovery of NiO), we see that the 1st step accounts for 84.16% of the total environmental impacts for the functional unit (1 g NiO and 2.02 g YSZ) on average. If we analyse the total value of the climate change, which is 2.134 kg CO₂eq. per 1 g NiO and 2.02 YSZ, we find that 62.6 % is due to the disaggregation step, 19.7 % to the recovery step and 15.5 % to the NaOH addition step. Other environmental impact indicators show similar relative contributions.

An additional analysis of electricity and chemical consumption for the EoL-SOC fuel electrode and electrolyte recovery process was performed to determine the contribution of chemical production and use as well as electricity consumption to the total environmental impacts (see

Fig. 8). The results show that, on average, 82.6 % of the total environmental impact is due to electricity consumption in the case of the EU electricity mix (EU grid mix 2020 [55]) and 17.4 % is due to the materials and chemicals used in the SOC fuel electrodes recovery process (mainly in the production of sodium hydroxide).

Sensitivity analyses were also carried out in the case of SOC fuel electrodes recycling process and three scenarios with different electricity mixes were defined: EU grid mix [55] – base case, French grid mix [60] and EU grid mix from renewable energy sources (RES grid mix [61]). The results are shown in relative comparison in Fig. 9, whereby the scenario with the highest impact is assigned 100% in the impact category.

The climate change indicator is reduced by 89% for the recycling process with the EU renewable energy grid mix and by 75% for the French grid mix, compared to the recycling process with the average EU grid mix. On the other hand, the resource use (fossil) is the highest for the French grid mix (69.89% nuclear) and the resource use (minerals and metals) increases almost fourfold for the RES grid mix compared to the EU grid mix, due to the renewable energy sources that consume many valuable minerals and metals.

Table 9
Results of the environmental impact indicators for recovered 1 g NiO and 2.02 g YSZ with the relative contribution of the individual EoL processes.

	EF 3.0 Acidification [Mole of H+ eq.]	EF 3.0 Climate Change [kg CO ₂ eq.]	EF 3.0 Eutrophication, freshwater [kg P eq.]	EF 3.0 Eutrophication, marine [kg N eq.]	EF 3.0 Eutrophication, terrestrial [Mole of N eq.]	EF 3.0 Resource use, fossils [MJ]	EF 3.0 Resource use, mineral and metals [kg Sb eq.]
Total NiO (1g) + YSZ (2.02g)	0.00443 (100%)	2.134 (100%)	7.8E-06 (100%)	0.00109 (100%)	0.0112 (100%)	43.56 (100%)	3.3E-07 (100%)
YSZ (2.02g)	84.1%	84.3%	83.2%	84.4%	84.2%	84.2%	84.7%
Mechanical Detachment (total)	0.6%	0.6%	0.8%	0.6%	0.6%	0.6%	0.6%
EU-28: Water (deionised) – M. detachment	0.0%	0.0%	0.2%	0.0%	0.0%	0.0%	0.0%
EU-28: Electricity grid mix - M. detachment	0.6%	0.6%	0.6%	0.6%	0.6%	0.6%	0.6%
Milling & Sieving (total)	1.4%	1.4%	1.4%	1.3%	1.4%	1.4%	1.3%
EU-28: Electricity grid mix - Milling & Sieving	1.4%	1.4%	1.4%	1.3%	1.4%	1.4%	1.3%
Disaggregation (total)	62.9%	62.6%	62.5%	61.2%	62.1%	63.1%	61.5%
EU-28: Water (deionised) – Disaggregation	0.0%	0.0%	0.2%	0.0%	0.0%	0.0%	0.0%
EU-28: Electricity grid mix – Disaggregation	62.9%	62.6%	62.3%	61.2%	62.1%	63.1%	61.5%
Recovery (total)	19.2%	19.7%	18.5%	21.3%	20.1%	19.1%	21.2%
EU-28: Electricity grid mix - Recovery	18.0%	18.0%	17.9%	17.6%	17.8%	18.1%	17.6%
DE: Nitric acid (98%) – Recovery	1.2%	1.7%	0.6%	3.7%	2.3%	1.0%	3.6%
NaOH addition + stirring (total)	15.7%	15.5%	15.5%	15.3%	15.6%	15.6%	15.1%
EU-28: Sodium hydroxide - NaOH addition + stirring	15.3%	15.2%	15.1%	14.8%	15.1%	15.3%	14.9%
EU-28: Electricity grid mix - NaOH addition + stirring	0.4%	0.4%	0.4%	0.5%	0.5%	0.3%	0.2%
Rinsing, Recovery, Drying (total)	0.1%	0.1%	1.2%	0.2%	0.2%	0.1%	0.1%
EU-28: Electricity grid mix - Rinsing, Recovery, Drying	0.1%	0.1%	0.1%	0.1%	0.1%	0.1%	0.1%
EU-28: Water (deionised) - Rinsing, Recovery, Drying	0.1%	0.1%	1.2%	0.1%	0.1%	0.1%	0.1%
Calcination (total)	0.1%	0.1%	0.1%	0.1%	0.1%	0.1%	0.1%
EU-28: Electricity grid mix - Calcination	0.1%	0.1%	0.1%	0.1%	0.1%	0.1%	0.1%

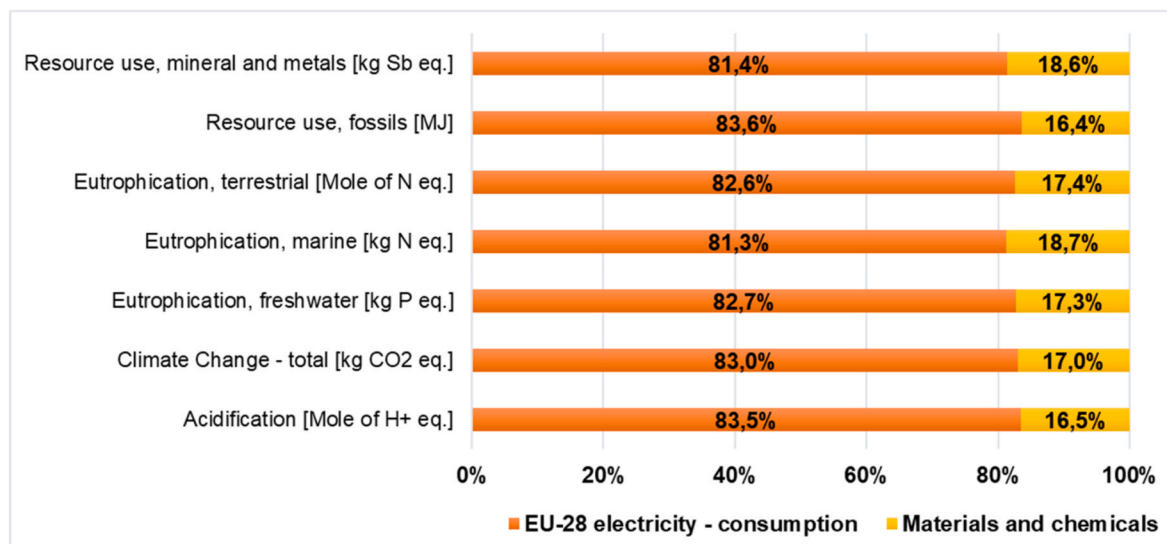


Fig. 8. Contribution to the total environmental impact due to electricity consumption (EU grid mix) and materials/chemicals used for 1 g NiO and 2.02 g YSZ recovery.

A reduction in energy consumption (EU electricity mix) of 30%, 50% and 70% is defined for three additional scenarios that represent an upscaling of the recycling process for SOC fuel electrodes. The relative reduction in impact categories is shown in Table 10, which shows that a 70% reduction in electricity consumption in the recovery process for 1 g

NiO and 2.02 g YSZ means an average reduction in impact categories of –57.8%.

The comparison of the results with similar studies dealing with recovery processes of SOC fuel electrode and electrolyte and oxygen electrodes is not yet possible due to the gap in the data and results

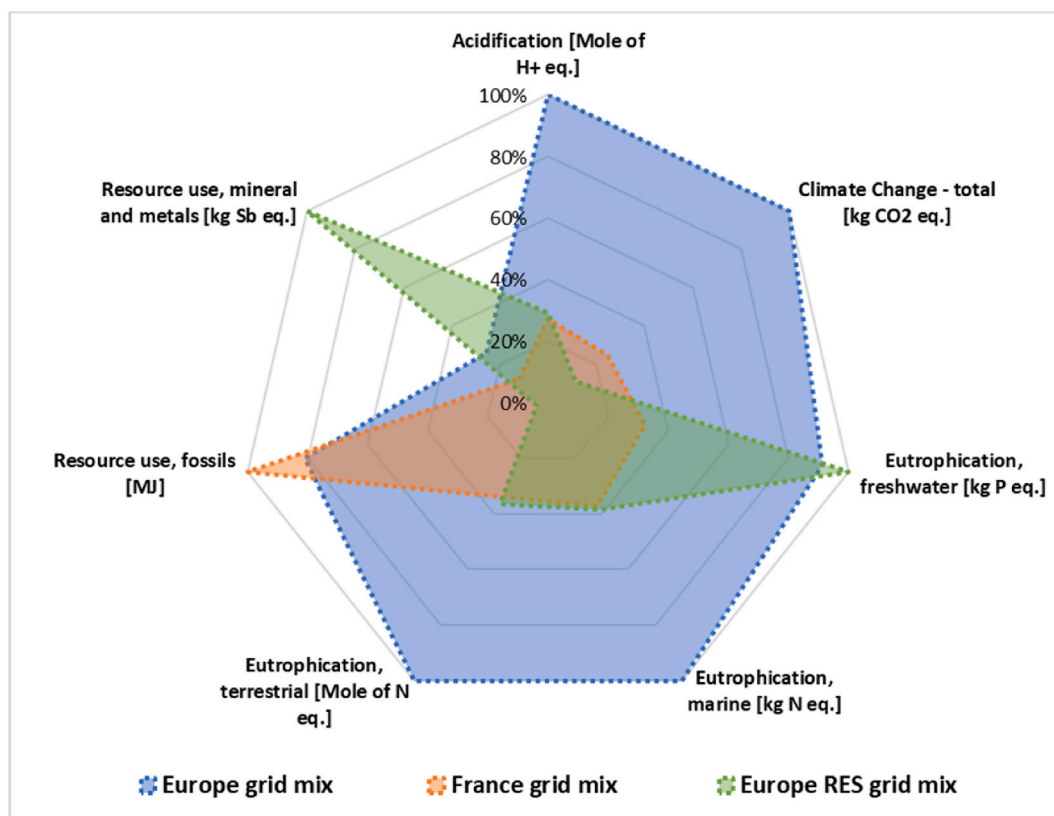


Fig. 9. Relative environmental impacts of for 1 g of NiO and 2.02 g of YSZ recycling processes with EU electricity (base case), France electricity and RES EU electricity grid mix.

Table 10

Total environmental impact reduction per FU with energy consumption reduction due to economy of scale.

	Total per FU 1 g NiO and 2.02 g YSZ	30% El. reduction	50% El. reduction	70% El. reduction
Acidification [Mole of H+ eq.]	100.0%	−25.0%	−41.7%	−58.4%
Climate Change-total [kg CO ₂ eq.]	100.0%	−24.9%	−41.5%	−58.1%
Eutrophication, freshwater [kg P eq.]	100.0%	−24.8%	−41.3%	−57.9%
Eutrophication, marine [kg N eq.]	100.0%	−24.4%	−40.6%	−56.9%
Eutrophication, terrestrial [Mole of N eq.]	100.0%	−24.8%	−41.3%	−57.8%
Resource use, fossils [MJ]	100.0%	−25.1%	−41.8%	−58.5%
Resource use, mineral and metals [kg Sb eq.]	100.0%	−24.4%	−40.7%	−57.0%

available in the scientific literature. Nevertheless, the recycling processes, when scaled up to demonstration and industrial levels, show the potential to have a significantly lower environmental impacts than virgin materials. At this stage in the development of recycling processes (TRL 3 – TRL 5), this is not yet feasible. The main reason for this is the high energy consumption due to the laboratory research level.

5. Conclusions

This study describes the development of life cycle inventories based on experimental results from laboratory-scale recycling processes (TRL3-TRL5) for solid oxide technology, based on work carried out as part of the EU BEST4Hy project. The newly developed life cycle inventories were used to calculate the environmental profile with an LCA study for the recovery process of lanthanum (as lanthanum oxide) and cobalt (as cobalt oxide) from the EoL SOC oxygen electrode and for the recovery process of YSZ and NiO from the EoL SOC fuel electrode and electrolyte. The study identifies and analyses the main hotspots within the recycling processes. As the electricity consumption and origin of electricity is one of the most important hotspots, a scenario analysis was

performed for different electricity consumptions (reduction of electricity consumption by upscaling to higher TRLs) and different electricity mixes (EU mix, nuclear energy, renewable energy mix). To show the impact of economy of scope.

The first part of the results presents the environmental impacts of the recovery of SOC oxygen electrode material for lanthanum oxide and cobalt oxide as the main output flows of the recycling process at TRL 3. In the case of lanthanum and cobalt recovery, the environmental impacts are evenly distributed across all sub-processes and there is no clear hotspot within the sub-processes. The overall climate change indicator for lanthanum oxide and cobalt oxide recovery is 3.638 kg CO₂ equivalents for 1.36 g of lanthanum oxide and 1 g of cobalt oxide recovered. The results also show that, on average, 66% of the total environmental impact is due to electricity consumption from the EU mix and 34% is due to the use of materials and chemicals. The scenario analysis of the electricity origin has shown that the climate change indicator can be reduced by 86% when using the EU mix from renewable energy compared to using the EU mix. To demonstrate the impact of future recycling processes on higher TRLs, scenario analyses with 30 %, 50 % and 70 % electricity reduction were analysed. The results show that

there is a great potential for a reduction in environmental impacts with an average reduction of –46% with a –70% reduction in energy use.

The LCA results for the SOC fuel electrode and electrolyte recovery at TRL 5 for the recovery of yttrium-stabilised zirconia and nickel oxide show that the main hotspot is the disaggregation process, which accounts for 62.27% of the total environmental impact on average. In the recycling process of YSZ and NiO, electricity is the main hotspot, accounting for an average of 82.6% of the total environmental impact. The total value of the climate change indicator is 2.134 kg CO₂-equivalents per 1 g of recovered NiO and 2.02 g of YSZ.

The present work is in the context of other work carried out in recent years in the field of LCA for solid oxide technologies, focusing on the EoL phase, where the largest gaps for LCA assessments exist. Based on the research presented, there is no other complete inventory of the recycling process of SOC in the literature. Therefore, the main added value of the present work is that for the first time a life cycle inventory for the recycling of critical SOC materials has been developed based on real experimental data. Hotspots have been identified and a critical evaluation has been performed, which will help to fill the missing gaps in the future development of life cycle inventories for SOC recycling technologies and can be used in potential future research related to SOC technologies.

CRedit authorship contribution statement

Mitja Mori: Conceptualization, Investigation, Project administration, Software, Supervision, Writing – original draft, Writing – review &

editing. **Jure Gramc:** Data curation, Methodology, Software, Validation. **Domen Hojkar:** Conceptualization, Formal analysis, Methodology and review. **Andrej Lotrič:** Conceptualization, Formal analysis, Methodology. **Federico Smeacetto:** Methodology, Project administration, Supervision, Writing – review & editing. **Sonia Fiorilli:** Methodology, Supervision, Validation, Writing – original draft. **Silvia Fiore:** Conceptualization, Formal analysis, Funding acquisition, Methodology, Project administration, Validation, Writing – original draft, Writing – review & editing. **Rok Stropnik:** Investigation, Methodology, Software, Validation, Writing – original draft, 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.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.ijhydene.2024.08.411>.

7 Abbreviations

ADP	Abiotic resource depletion
AE	Accumulated Exceedance
CCHPS	Combined cooling, heating, power, and storage
CHP	Combined heat and power
Co ₃ O ₄	Cobalt oxide
CRMs	Critical raw materials
DE	Germany
EF3.0	Environmental Footprint 3.0
EoL	End-of-Life
EU	European Union
FCH	Fuel Cells and Hydrogen technologies
GDC	Cerium gadolinium oxide (20% GDC)
GWP100	Global Warming Potential (100 years)
H ₂ O ₂	Hydrogen peroxide
HNO ₃	Nitric acid
HT	high temperature
La ₂ O ₃	lanthanum oxide
LCA	Life Cycle Assessment
LCI	Life Cycle Inventory
LCIA	Life Cycle Impact Assessment
LSC	Lanthanum Strontium Cobaltite
LSC64	Lanthanum Strontium-doped Cobaltite (La _{0.6} Sr _{0.4} CoO ₃)
NaOH	Sodium hydroxide
Ni	nickel
NiO	nickel oxide
Ni-YSZ	Nickel–Ytria Stabilised Zirconia
REC	Recycling
RER	region Europe
RES	Renewable energy source
SOCs	Solid Oxide Cells
SOECs	Solid Oxide Electrolysis Cells
SOFCS	Solid Oxide Fuel Cells
SS	Stainless steel
TRL	Technology readiness levels
YSZ	Yttrium Stabilised Zirconium

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