Critical materials in PEMFC systems and a LCA analysis for the potential reduction of environmental impacts with EoL strategies

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Abstract
Commonly used materials constituting the core components of polymer electrolyte membrane fuel cells (PEMFCs), including the balance-of-plant, were classified according to the EU criticality methodology with an additional assessment of hazardousness and price. A life-cycle assessment (LCA) of the materials potentially present in PEMFC systems was performed for 1 g of each material. To demonstrate the importance of appropriate actions at the end of life (EoL) for critical materials, a LCA study of the whole life cycle for a 1-kW PEMFC system and 20,000 operating hours was performed. In addition to the manufacturing phase, four different scenarios of hydrogen production were analyzed. In the EoL phase, recycling was used as a primary strategy, with energy extraction and landfill as the second and third. The environmental impacts for 1 g of material show that platinum group metals and precious metals have by far the largest environmental impact; therefore, it is necessary to pay special attention to these materials in the EoL phase. The LCA results for the 1-kW PEMFC system show that in the manufacturing phase the major environmental impacts come from the fuel cell stack, where the majority of the critical materials are used. Analysis shows that only 0.75 g of platinum in the manufacturing phase contributes, on average, 60% of the total environmental impacts of the manufacturing phase. In the operating phase, environmentally sounder scenarios are the hydrogen production with water electrolysis using hydroelectricity and natural gas reforming. These two scenarios have lower absolute values for the environmental impact indicators, on average, compared with the manufacturing phase of the 1-kW PEMFC system. With proper recycling strategies in the EoL phase for each material, and by paying a lot of attention to the critical materials, the environmental impacts could be reduced, on average, by 37.3% for the manufacturing phase and 23.7% for the entire life cycle of the 1-kW PEMFC system.

KEYWORDS
critical materials, end of life, life-cycle assessment, PEM fuel cells, recycling
INTRODUCTION

It is expected that the world's primary energy mix in 2050 will be divided equally between fossil and nonfossil sources in the two main energy sectors: electricity/power and transportation. In the more specific market for electricity generation with renewable energy sources (RESS), solar photovoltaic and wind will likely achieve shares of 40% and 29%, respectively. This introduction of unpredictable RESs into the electricity-grid mix will be closely linked to the introduction of energy storage for balancing the electricity grid. Fuel cell and hydrogen (FCH) technologies could play an important role in energy storage and grid balancing between producers and the consumers. In addition to the energy generation sector, the increase in the use of hydrogen as an energy carrier for fuel cell electric vehicles (FCEVs) or in industry as a green and sustainable product for industrial uses is the main driver toward a future hydrogen economy.

In this paper, the technology under consideration is the polymer electrolyte membrane fuel cell (PEMFC). The materials of interest as part of FCH technologies must be selected and evaluated with a multi-criteria tool, due to their different impacts on the environment, human health, the economy, etc. From the sustainability point of view, it is necessary to achieve a reduction in the use of these critical materials and consequently reduce the environmental impacts during the manufacturing phase. In the past decade, the proposed strategies from the National Research Council and the European Commission in 2008 have accelerated the development of methods for assessing the criticality of raw materials. As a result, numerous reports using existing methodologies were published: Material scarcity by Wouters and Boi, Critical metals for future sustainable technologies by Buchert al, Risk List by British Geological survey, Assessment of critical minerals by the National Science and Technology Council, and Methodology for Establishing the EU List of Critical Raw Materials by Blengini et al. Additional reports based on existing methodologies were published in scientific papers by Glöser et al., Bertrand et al., McCullough et al., Daw et al., and Blengini et al. Others, such as Wagner et al., Knoeri et al. and Weiser et al. proposed new methodologies for critical material assessments. The European Union (EU) is following on from its Raw Materials Initiative by funding various research projects that are concerned with increasing the supply or reducing the demand of at least one critical material.

Additional information on the criticality of a material (from the environmental point of view) can be obtained by using a life-cycle assessment (LCA). Within the Fuel Cells and Hydrogen Joint Undertaking (FCHJU), recommendations and guidelines for carrying out a LCA of FCH technologies were published by the FC-HyGuide project. A LCA study is divided into several steps, with most of them conducted sequentially, but there are also iterative parts where the previous steps have to be reconsidered. Each step is subdivided into several categories, which need to be properly addressed in order to ensure a proper LCA. There are recent LCA studies on PEMFC systems that have a detailed description of the LCA (following the guidelines of the FC-HyGuide project), a list of materials comprising the systems under consideration, and some of them also include the end-of-life (EoL) phase. The LCA of a cargo bike fitted with a PEMFC stack was performed by Garrain and Lechon. The goal of the study was to identify the major impacts related to the manufacturing and disposal phases, while the operating phase was not considered. The results showed that platinum has a high acidification impact, due to the extraction of the material. However, copper is the main contributor to acidification as a result of its high mass percentage. Similarly, a LCA of a PEMFC’s membrane electrode assembly (MEA), where only the production and the EoL phases were considered, was performed by Laforest et al. The two EoL scenarios were compared: the first scenario assumed only an incineration process and the second also included the recycling of the platinum and gasket materials. The assessment revealed that the MEA life-cycle impact can be reduced by 60% if the electrode is recycled at the EoL phase. In addition, the main impact category decreases in proportion to the platinum recycling rate. A LCA and comparison of an internal combustion engine vehicle (ICEV), a battery electric vehicle (BEV), and a FCEV were made by Evangelisti et al. The study analyzed the whole life cycle, including the manufacturing of the vehicle, its use phase and the EoL. In the case of the FCEV, the PEMFC stack and all the balance-of-plant (BoP) components, including the hydrogen tank, were included in the LCA study. The global warming potential (GWP) of the disposal phase was shown to be negligible for all three technologies. The benefit of using hydrogen in the FCEV was shown in a reduced GWP for the use phase, when compared with the other two technologies. However, reducing the environmental impact associated with the manufacture of a FCEV still represents an important challenge that needs to be addressed. The objective of the LCA study performed by Di Marzobardino et al. was to evaluate the environmental burdens of a commercial PEMFC micro-CHP system with a natural gas steam reformer, a Reformcell micro-CHP system with an auto-thermal reformer, and a hydrogen-selective membrane reactor (ATR-MR) over their whole life cycles. The results showed the advantages of the ATR-MR system in terms of carbon footprint, resources, and water withdrawal as a consequence of a higher electrical efficiency. For the ecosystem quality and human health indicators, the ATR-MR showed a slightly higher impact for some of the scenarios studied, compared with the natural gas steam reformer system. The direct emissions from the reformer and the auxiliary boiler only have an influence on the greenhouse gas emissions, for which they...
are one of the largest contributors. The production, maintenance, and EoL of the micro-CHP are, in global terms, small contributors to the impacts, except for human health. An uninterrupted-power-supply (UPS) system based on a PEMFC was studied throughout its whole life cycle by Stropnik et al.27 Three EoL scenarios were assumed, where the base case scenario assumes only a landfilling process, the feasible scenario was based on the greatest possible theoretical recycling and reuse possibilities without landfilling, and the realistic scenario included all three processes. The results showed that the EoL phase has a lower environmental impact compared with the other phases. However, with a circular economy (re-cycling and reuse of the materials in the EoL phase) an average reduction of 66% for all environmental impact indicators could be achieved for the entire life cycle of an UPS system operated in Norway.

The present paper is based on research conducted within the HyTechCycling project,28 which is one of the projects funded by FCH JU (GA700190). In the first part, the materials that can be used in PEMFC systems are assessed according to the EU criticality methodology,12,29 with additional assessments of hazardosity and price. The PEMFC system’s materials are further assessed with a LCA methodology, which provided additional information about their environmental impacts for the production phase. In the second part, the LCA case study on a 1-kW PEMFC system is performed. The material masses and electricity consumption in the manufacturing phase are taken from the literature, and for the operating phase, the available data from manufacturers are used. Also, four different scenarios of hydrogen production are assumed. The main contribution of this research is the analysis and modeling of the EoL phase, where detailed modeling of the Pt hydrometallurgical recycling from Duclos et al.30 is included. Finally, the potential for the reduction in environmental impacts through various EoL strategies for reducing the use of virgin materials or energy is analyzed.

2 POLYMER ELECTROLYTE MEMBRANE FUEL CELLS

The main components of a PEMFC are the polymer electrolyte membrane (PEM) and two electrodes with an applied catalyst layer (Figure 1). The two electrodes (called the anode and the cathode) are electrically insulated and separated by the PEM, which also serves as a barrier for the reactant gases, but allows the protons to migrate across it. These three components are often referred to together as the membrane electrode assembly (MEA). On the anode side, a hydrogen oxidation reaction (HOR) takes place where the electrons are separated from the protons. The separated electrons travel via the external electron-conductive circuit, through an electrical load, where the generated electrical output of the fuel cell is used, and the protons pass through the PEM to the cathode side of the fuel cell. On the cathode side, where the oxygen reduction reaction (ORR) takes place, the protons and electrons combine with oxygen to produce water. The main electrochemical reactions take place on the triple phase boundary where the electrolyte, the catalyst, and the reactant are all in contact (see Figure 1).

The gases flow through the flow-field channels embedded in the bipolar plates. The bipolar plates are located on each side of the MEA, and besides distributing the gases, they must also be highly electrically conductive and serve as current collectors. The bipolar plates are usually made from graphite, graphite composites, or stainless-steel materials. On the electrode, the gas diffusion layer (GDL) is usually made from carbon paper or cloth and the catalyst layer is made of Pt or a Pt-alloy catalyst supported on carbonaceous material. The PEM is made from sulfonated polymers that are highly proton conductive and have hydrophilic properties (they contain liquid water). The most common commercially available PEMs (eg, Nafion® from DuPont) are made of perfluorosulfonic acid (PFSA). However, recently, as a lower-cost alternative, sulfonated polyether ether ketone (S-PEEK) and poly styrene sulfonic acid (PSSA) are also used.31 Additionally, some types are based on solid proton conductors, for example, zirconium phosphates.32,33 In this paper, the Nafion® membrane was assumed in the case studies. The Nafion® PFSA membrane is produced via the copolymerization of an unsaturated perfluoralkyl sulfonyl fluoride (PSF) with tetrafluoroethylene (TFE). The relative amount of TFE to PSF is approximated by a 6:1 molecular ratio, which works out at 57.4 wt.% TFE to 42.6 wt.% PSF.34

The typical operating conditions of a PEMFC are absolute pressure from ambient to 3 bar and temperatures between 45 and 85°C (for a low-temperature PEMFC). A special type of PEMFC, known as a high-temperature PEMFC (up to 200°C31), can operate at higher temperatures, where a mineral acid-based electrolyte, such as polybenzimidazole (PBI) doped with H3PO4, and less-expensive composite membranes35 are used.

3 CLASSIFICATION METHODOLOGY OF THE MATERIALS

One of the goals of the paper is to identify the critical materials according to the classification methodology used in the HyTechCycling project.36 In this project, the considered FCH technologies were broken down into the main components and the most common materials comprising them were identified. The three main criteria for the material classification in the FCH technologies were hazardousness, EU criticality methodology, and price. All these classification criteria
are described in detail in the publicly available deliverable D2.1\textsuperscript{37} of the HyTechCycling project.

Hazardous materials are those which are dangerous or have a harmful effect on human health or the environment. Hazardousness in D2.1 of the HyTechCycling project was assessed according to the Priority List of Toxic Substances\textsuperscript{38} and the Handbook on the Toxicology of Metals.\textsuperscript{39} To determine the scarcity of the FCH materials, the EU criticality methodology was used with two assessment criteria\textsuperscript{40}: economic importance (EI) and supply risk (SR). In Table 1, the critical materials are presented according to the EU commission and EU criticality methodology.

**FIGURE 1** Operating principle of a PEMFC

| **2017 Critical Raw Materials (27)** |
|-------------------------------------|
| **2017 EU critical raw materials list**\textsuperscript{40} |
| Antimony | Fluorspar | LREEs | Phosphorus |
| Baryte | Gallium | Magnesium | Scandium |
| Beryllium | Germanium | Natural graphite | Silicon metal |
| Bismuth | Hafnium | Natural rubber | Tantalum |
| Borate | Helium | Niobium | Tungsten |
| Cobalt | HREEs | PGMs | Vanadium |
| Coking coal | Indium | Phosphate rock |
All raw materials, even when not classed as critical, might still be important for the EU economy. Therefore, according to this assessment a material is marked as High in EU criticality if it is present in the 2017 EU CRM’s list (exceeds EI and SR threshold). If a material exceeds the threshold only in one assessment criterion, the material is marked as Medium; in all other cases, the material is marked as Low in the EU criticality methodology.

The prices of the materials used in the PEMFC were estimated according to the Asian Metal Market, the London Metal Exchange, and the Elements and their Compounds list. The materials were categorized according to price as: Low (<5 $/kg), Medium (5-500 $/kg), and High (>500 $/kg).

Based on the presented methodology, the lists of all the materials that could be used in the PEMFC system during the manufacturing phase were composed. The material is marked as critical if it is classified as follows: hazardous, high in price, high in EU criticality, or medium in both EU criticality and price. Critical material means that it has a substantial potential to reduce the environmental impact during the manufacturing phase if the appropriate EoL strategies are applied.

The critical BoP (Table 2) materials are elastomer silicone, Pd, and PTFE due to their high EU criticality. The noble metals (Au, Ag) are critical due to their high price. Batteries (LiFePO4), Sn, Pb, and Pd are critical due to their toxicity or combination of price and EU criticality.

### Table 2: List of common materials in PEMFC systems

| Material                        | Component (Subsystem)                      | Hazardous | Price | EU criticality |
|---------------------------------|--------------------------------------------|-----------|-------|----------------|
| Aluminum                        | Housing (BoP, Stack)                       | No        | Low   | Medium         |
| Carbon                          | GDL (Stack)                                | No        | Low   | Low            |
| Chromium steel                  | Housing (BoP)                              | No        | Low   | Medium         |
| Copper                          | PCB’s (BoP)                                | No        | Low   | Low            |
| Elastomer (EPDM)                | Sealing (BoP, Stack)                       | No        | Low   | Low            |
| Elastomer (Viton)               | Sealing (BoP, Stack)                       | No        | Medium| Low            |
| Elastomer (Silicone)            | Sealant (BoP, Stack)                       | No        | Low   | High           |
| Glass wool                      | Insulation (BoP, Stack)                    | No        | Low   | Low            |
| Gold (Au)                       | Ancillary BoP (BoP)                        | No        | High  | Low            |
| Graphite                        | GDL, Interconnect (Stack)                  | No        | Low   | Medium         |
| Plastics (HDPE, PP, Nylon, PVdC)| Ancillary BoP (BoP, Stack)                 | No        | Low   | Low            |
| Lead (Pb)                       | Batteries (BoP)                            | Yes       | Low   | Medium         |
| Lithium-ion (LiFePO4)           | Batteries (BoP)                            | No        | Medium| Medium         |
| Palladium (Pd)                  | Catalyst, Ancillary BoP (BoP, Stack)       | Yes       | High  | High           |
| Perfluorosulfonic acid (PFSA) - Nafion | Electrolyte (Stack)                   | No        | Medium| Medium         |
| Platinum (Pt)                   | Catalyst (Stack)                           | No        | High  | High           |
| Polybenzimidazole (PBI) doped with H3PO4 | Electrolyte (HT-Stack)                  | Yes       | Medium| High           |
| Polystyrene sulfonic acid (PSSA)| Electrolyte (Stack)                        | No        | Low   | Medium         |
| Polytetrafluoroethylene (PTFE  – Teflon) | Catalyst, Sealant, Ancillary BoP, GDL (BoP, Stack) | No | Medium | High |
| Ruthenium                       | Catalyst (Stack)                           | Yes       | High  | High           |
| Silver (Ag)                     | Ancillary BoP (BoP)                        | No        | High  | Medium         |
| Steel products, Steel alloys    | Housing, GDL, Interconnect (BoP, Stack)    | No        | Low   | Medium         |
| Sulfonated polyether ketone (s-PEEK) | Electrolyte (Stack)                   | No        | Medium| Medium         |
| Tin (Sn)                        | Batteries (BoP)                            | Yes       | Medium| Medium         |

A list of commonly used materials in PEMFC systems and assessments of these materials are in this table and critical materials in PEMFC technologies are highlighted (Bolded). Bold text showing critical materials, which are important to take care in EoL.
The materials in PEMFC systems are mainly low to medium in terms of cost, with the exception of the PGMs (Pt, Ru, Pd). The majority of the materials used in this FCH technology are classified as nonhazardous. The most critical materials in the PEMFC technology are PGMs and PEMs made from PFSA, or in the case of high-temperature PEMFC polybenzimidazole (PBI), doped with H$_3$PO$_4$. Other materials are critical due to their combination of price and EU criticality.

4 | LIFE-CYCLE ASSESSMENT APPROACH

The life-cycle assessment (LCA) methodology is standardized according to ISO standards 14040 and 14044 and is used to analyze the environmental impact of the entire life cycle of products or activities. Provisions and suggestions from the FC-HyGuide project for performing an LCA on FCH technologies were also considered in this analysis. For this purpose, GaBi Thinkstep software was used.

4.1 | Life-cycle assessment of materials in the production phase

Materials that could be used in the manufacture of PEMFC technologies are listed in Table 2.

The preliminary LCA of the materials used in the PEMFC stack and the BoP was based on a unit of mass (1 gram of each material) and performed using the generic data from Ecoinvent 3.3, 3.5 and the Gabi professional database (ts). The materials that are present in the LCA model of the PEMFC media are not necessarily used by all manufacturers, but they were identified as possible to use.

4.2 | Life-cycle assessment of PEMFC system

A life-cycle inventory (LCI) analysis and the data needed for modeling the life-cycle phases were gathered from a previous EU project and the literature with the help of data sheets from FC manufacturers that are publicly available. The main goal of this study was to analyze the environmental impacts of manufacturing, operating, and the EoL phase for a 1-kWe PEMFC system and to discuss the importance of proper EoL strategies. In the EoL phase, a realistic strategy of materials recycling was modeled to assess the potential reduction of environmental impacts in the PEMFC life cycle. A 10-kWe PEMFC system was taken as a case study for the material inputs, but all the data were recalculated and normalized to the unit of 1 kW of electric power, which makes the LCA model more general. The functional unit is defined as 1 kWh of energy, which means 1 kWh of produced electricity. The life-cycle impact assessment (LCIA) methodology used in the analysis was the midpoint-approach method CML2001, with an additional comparison using Environmental Footprint 2.0 in Section 5.2.3. The scope of the study was from “cradle to grave,” with the emphasis on the potential of proper EoL strategies. Conventional materials such as steel, plastic, aluminum were recycled according to the industry data and modeled with the help of generic databases. Critical materials such as Pt, for which standard recycling industry processes are not set up, were recycled according to Duclos et al. The Gabi Thinkstep software environment was used to set up the LCA models.

The LCA model includes all the materials that could be present in the PEMFC technologies. However, the LCA model shown in Figure 2 contains a limited list of materials, which is shown in Table 3. The data used for modeling the manufacturing phase of the 1-kWe PEMFC stack were taken from an FP7 EU project and for the operating phase, publicly available datasheets from Needstack were used. In the EoL phase, recycling was used in most cases with specific recycling rates regarding the process and the materials used. Energy extraction and landfill were used just in cases where reuse or recycling was not possible.

4.2.1 | Manufacturing phase

The masses of the materials and the energy input in the form of electricity are presented in Table 3. The data were recalculated in accordance with the 1-kWe system of the PEMFC. The electricity needed to produce the PEMFC system is presumed to be generated from the EU-28 electricity mix, but the model is set up in such a way that it is possible to change all the input materials’ masses and the type of electricity mix that normally corresponds to the country where the manufacturer’s facilities are located. All the materials in the case of the LCA model are virgin materials. In this case, the EoL phase offers the possibility to reduce the environmental impacts in the manufacturing phase with specific material recycling/recovery processes.

4.2.2 | Operating phase

The operating phase of a PEMFC represents conversion of the chemical energy of hydrogen into electricity. Hydrogen is an energy vector, which means it is not a naturally occurring energy source; therefore, it has to be produced. According to several recent reports and papers, hydrogen is produced worldwide from several sources, but fossil fuels are responsible for 96%. Natural gas steam reforming (NG-SR) holds the largest share of the production from hydrocarbon sources at 48%; the second largest part is oil-based at 30%; and 18% is derived from coal gasification; while the electrolysis of water is responsible for the remaining 4%. It is important to note that all these reports and papers refer to various sources that ultimately cite the original sources, which are from the previous...
decade or even older. Examples of original sources are the NREL report from 1999,65 and the NRDC reports from 200466 and 2008.67 Nevertheless, all the original researches show very similar shares of worldwide hydrogen production technologies. Since NG‐SR is the most economic and efficient method of producing hydrogen,68 this is currently the predominant technology. In the USA, around 95% of all hydrogen is produced with NG‐SR.68 However, in order to increase the share of renewable hydrogen production, it is necessary to increase the share of electrolysis from RESs.

In this research, only two hydrogen production methods are presumed: a) water electrolysis and b) NG‐SR. The electricity needed for electrolysis (189 MJ/kg of H2) is generated from three different energy‐grid mixes, which are EU Hydro power, SI Lignite, and the EU‐28 mix, from the Gabi Professional Database.49 Altogether, this makes four different scenarios in the operating phase. The EU28 electricity mix presents (scenario 1—EU mix) the basic approach of hydrogen production with electrolysis; the EU28 hydro mix (scenario 2—EU hydro) represents the best‐possible or future RES‐based hydrogen production with electrolysis; and SI lignite (scenario 3—SI lignite) as the worst electrolysis hydrogen production scenario. On the other hand, the majority of hydrogen today is produced with NG‐SR; therefore, this scenario (scenario 4—NG reforming) is also evaluated in the operating phase.

For the operating lifetime of a stationary PEMFC system, 20 000 h was chosen, with no need to replace the stack in this timeframe.53 The maximum hydrogen flow is 150 Nl/min where, according to the National Institute of Standards and Technology (NIST), normal conditions are taken to be at a pressure of 1 atm and a temperature of 20°C. In the whole operating lifetime of 1kW PEMFC that corresponds to 1507.6 kg of hydrogen consumption, which is produced by electrolysis or NG‐SR. If the hydrogen is produced with electrolysis, that corresponds to 284,855.6 MJ of electricity consumption.

4.2.3 | End‐of‐Life phase

In the EoL phase, different approaches were used for the PEMFC stack materials: recycling (REC), energy extraction (EE), and landfill (LF), as a worst‐case scenario, where no other data were available for the EoL.22 The first step in the EoL phase was manual dismantling of the system, where process treatment of the used industrial electronic device, manual dismantling from Ecoinvent 3.5, was used. In Table 4, the materials inputs for the EoL processes and the material‐recovery ratio are presented for a 1‐kW PEMFC system. For each material, the EoL approach, recycling ratio, and the used EoL process from the databases are shown. The EoL processes from the database were scaled according to the inventory data. The recycling ratio is defined according to the literature, industry data, or research papers.30,69-71

After the recycling process, secondary materials are available (Table 5) that can be used in the PEMFC system's manufacturing phase or in the manufacturing processes of other technologies. The reduced environmental impacts due to avoided (secondary) materials are subtracted from the environmental impacts in the manufacturing phase of the PEMFC unit.
For the recycling of platinum, which is one of the most critical materials in PEMFC technology, the process was modeled according to a recent publication about Pt recovery with a hydrometallurgical process by Duclos et al. The LCA model of the process is presented in Figure 3, where the mass and energy balances are set according to the output of 1 kg of Pt. The masses and energy demands for Pt recycling in the observed PEMFC system were scaled to an output of 0.00057 kg of Pt. This is the mass of secondary Pt that, according to Duclos et al., could be obtained with a 76% recycling ratio from the MEA. The remaining solid waste was incinerated as hazardous waste. Other solvents needed in the process (e.g., NaOH, demi water, H₂O₂, NH₄Cl) were modeled with a wastewater treatment after the recycling process. The hydrochloric acid needed for the recycling process should be neutralized after use; however, since the neutralization process is not available in the databases, it was not included in the LCA model.

Aluminum was recycled according to the EEA data available in the GaBi professional ts database. Steel recycling was modeled with the use of a cast iron production process and the mass allocation of environmental impacts according to the amount of steel scrap used in the process. Polypropylene (PP) and high density polyethylene (HDPE) were recycled using a plastic granulate secondary process. Solid waste (e.g., graphite, glass fibers, and Nafion® membrane) was incinerated using hazardous waste. In the incineration process for PVdC, the energy is extracted in the form of electricity (2.87 MJ) and heat (5.24 MJ), which could be used in the manufacturing phase of the PEMFC system. Therefore, the energy from the incineration process is subtracted from the overall environmental impacts in the manufacturing phase.

## RESULTS AND DISCUSSION

### 5.1 Life-cycle assessment of 1 gram of the materials in PEMFC technologies

Environmental indicators according to CML2001 are presented in Table 6 for the BoP and in Table 7 for the PEMFC stack materials. The analyzed environmental indicators in Tables 6 and 7 are as follows: Abiotic depletion potential—Elements (ADP elements), Abiotic depletion potential—Fossil fuels (ADP fossil), Acidification potential (AP), Eutrophication potential (EP), Freshwater aquatic ecotoxicity potential (FAETP), Global warming potential (100 years) (GWP 100), Human toxicity potential (HTP), Marine aquatic ecotoxicity potential (MAETP), Ozone depletion potential (ODP), Photochemical ozone creation potential (POCP), and Terrestrial ecotoxicity potential (TETP).
The environmental impact of each material is shown, regardless of the mass ratio that is usually defined according to the manufacturing data or inventory. With the described approach, this study wants to stress the basic impacts of the materials per unit of mass, in this case 1 gram. The green-colored cells represent low, the yellow cells represent mid and the red cells represent a high environmental impact in the selected environmental impact indicator of the CML2001 methodology.

In the BoP components (Table 6), gold is the material with the largest environmental impact in all the observed impact categories. Palladium has, on average, a one-order-of-magnitude lower environmental impact than gold, with silver having two magnitudes lower, while tin, nickel, and PFTE have already a three-magnitudes-lower impact. The lowest environmental impacts in the BoP materials come from plastics, aluminum, steel, etc.

In the case of PEMFC stack materials (Table 7), there is no gold present, but there are PGMs, which are very important materials for the EU economy. They are also important from the geostrategic point of view, and they are classified as critical (Table 2). Platinum has the largest environmental impact of all the potentially used materials in PEMFC core technologies, with palladium being, on average, one-order-of-magnitude lower, and ruthenium with a two-magnitudes-lower environmental impact.

Materials with the lowest impact are plastics, metals, and graphite, with Nafion and carbon black being in the middle, which is still at least a five-magnitudes-lower impact per 1 gram of manufactured material compared with PGMs.

Results are not usually presented in this form, since the mass ratios of individual materials are entirely different in the real components. However, this is still a very important basis for understanding the environmental impacts of the BoP and PEMFC technologies in the production phase.

From the results in Tables 6 and 7, it can be concluded that the most critical materials from the environmental point of view are the PGMs (Pt, Pd, and Ru) and precious metals (Au, Ag), followed by other metals (eg, Tin, Copper). Some parallels can be drawn between these results and the methodology for critical materials described in Section 3. All the PGMs and precious metals are critical according to the assessment methodology, due to their high price (eg, Au) or...
high EU criticality (Pt). Among these materials (PGMs and precious metals), the most important and most commonly used material in PEMFC technology is Pt, which has a significant environmental impact during the manufacturing phase. Therefore, it is necessary to reduce the quantity in the scope of the manufacturing technology and consider the EoL phase.

5.2 Life-cycle assessment of a 1-kW PEMFC system

5.2.1 Environmental impacts of the manufacturing phase

In the case of the manufacturing phase of the 1-kW PEMFC system, data from an EU project were used, which serve as a good example. The data for the manufacturing phase of a 1-kWe PEMFC system are presented in Table 3 and the LCA model with material flows in Figure 2. The absolute values for the manufacturing phase of a 1-kW PEMFC system are presented in Table 8, with the contribution of the PEMFC stack, the BoP, and the electricity consumption for manufacturing. The radar diagram in Figure 4 shows the contribution of the PEMFC stack, the BoP and the electricity consumption to the overall environmental impacts of the manufacturing phase of a 1-kWe PEMFC system.

In other technologies/products, electricity is usually the main contributor to the overall environmental impacts, but not in the case of hydrogen technologies, where the main environmental impact (9 out of 11) in the manufacturing phase comes from the core components in the PEMFC stack, where

![FIGURE 3 Life-cycle assessment model of Pt recycling process with boundary conditions](image)

### TABLE 6 Absolute values of environmental indicators for potential BoP component materials (per 1 gram of material)

| Material      | ADP elements [kg Sb eq.] | ADP fossil [MJ] | AP [kg SO₂ eq.] | EP [kg phosphate eq.] | FAETP inf. [kg DCB eq.] | GWP 100 [kg CO₂ eq.] | HTP inf. [kg phosphate eq.] | MAETP inf. [kg DCB eq.] | ODP steady state [kg R11 eq.] | POCP [kg Ethene eq.] | TETP inf. [kg DCB eq.] |
|---------------|--------------------------|-----------------|-----------------|------------------------|--------------------------|----------------------|---------------------------|--------------------------|-----------------------------|----------------------|-----------------------|
| LiFePO₄        | 3.80E-08                 | 1.53E-01        | 4.35E-05        | 5.27E-06               | 6.51E-05                 | 1.02E-02             | 8.64E-04                 | 7.62E-01                 | 1.25E-13                    | 1.01E-06             | 7.51E-05              |
| HDPE          | 4.17E-10                 | 6.43E-02        | 2.07E-06        | 2.32E-06               | 3.25E-06                 | 1.21E-02             | 4.36E-04                 | 7.87E-01                 | 6.06E-10                    | 2.97E-06             | 9.78E-06              |
| PTFE          | 1.60E-06                 | 2.04E-01        | 4.06E-05        | 8.26E-05               | 4.10E-02                 | 1.97E-01             | 1.00E-14                 | 2.17E-06                 | 1.82E-05                    | 1.01E-16             | 5.06E-07              |
| Aluminium     | 4.11E-09                 | 8.95E-02        | 4.01E-05        | 6.08E-05               | 3.79E-05                 | 3.74E-05             | 2.85E-02                 | 1.91E-02                 | 2.4E-01                     | 1.66E-05             | 8.64E-04              |
| Silicone      | 2.07E-07                 | 8.93E-02        | 1.68E-05        | 5.85E-05               | 1.17E-03                 | 6.31E-01             | 7.47E-15                 | 1.82E-06                 | 9.38E-04                    | 2.12E-02             | 1.82E-05              |
| Copper        | 1.81E-06                 | 3.97E-02        | 3.69E-04        | 7.68E-02               | 2.04E-02                 | 2.64E-01             | 2.04E-02                 | 1.16E-05                 | 9.78E-04                    | 1.01E-16             | 8.64E-04              |
| Gold          | 5.56E-02                 | 1.87E-02        | 1.56E-06        | 1.57E-09               | 3.98E-02                 | 1.59E-01             | 5.77E-02                 | 1.13E-06                 | 1.62E-06                    | 1.21E-02             | 1.25E-05              |
| Lead          | 2.78E-06                 | 1.15E-02        | 3.09E-05        | 7.14E-06               | 1.74E-03                 | 1.23E-02             | 3.44E-03                 | 4.86E-00                 | 1.69E-10                    | 1.57E-06             | 1.16E-05              |
| Chromium steel| 3.23E-08                 | 2.73E-02        | 1.58E-05        | 5.84E-06               | 6.04E-03                 | 2.86E-03             | 1.89E-02                 | 1.69E-01                 | 1.41E-10                    | 1.42E-06             | 6.12E-04              |
| Steel product | 9.39E-09                 | 2.09E-02        | 1.13E-05        | 5.14E-06               | 3.47E-03                 | 2.19E-03             | 2.84E-03                 | 1.45E-01                 | 1.11E-10                    | 1.98E-05             | 6.09E-05              |
| Palladium     | 5.37E-04                 | 5.74E-01        | 1.80E+00        | 3.79E-02               | 1.68E+01                 | 5.12E+00             | 1.43E+01                 | 3.37E+04                 | 2.38E-07                    | 7.25E-02             | 2.85E-02              |
| Silver        | 5.07E-04                 | 3.69E+00        | 3.25E-03        | 5.55E-03               | 1.29E+00                 | 3.35E-01             | 1.90E+00                 | 3.94E+03                 | 2.73E-08                    | 2.58E-04             | 1.94E-03              |
| PP            | 5.30E-11                 | 6.66E-02        | 6.23E-06        | 7.44E-07               | 5.57E-07                 | 2.00E-03             | 9.20E-06                 | 6.09E-02                 | 2.90E-16                    | 9.27E-07             | 5.87E-08              |
| Tin           | 1.90E-05                 | 2.53E-02        | 5.15E-04        | 5.54E-05               | 7.55E-03                 | 2.38E-02             | 1.00E-02                 | 2.99E+01                 | 1.52E-09                    | 2.59E-05             | 7.33E-05              |

The green-colored cells represent low, the yellow cells represent mid and the red cells represent a high environmental impact in the selected environmental impact indicator of the CML2001 methodology.
the most critical materials are used. The exceptions are the TETP and HTP indicators, where the main environmental impacts come from the manufacturing of the BoP components: this is due to the larger amount (1.1 kg—from Table 4) of chromium steel compared with the PEMFC stack (0.1 kg). In order to analyze and assess which materials have the largest environmental impact in the manufacturing phase of a 1-kWe PEMFC system, the results of the environmental balances of the PEMFC and the BoP LCA model have to be analyzed (Tables 9 and 10).

The environmental impact of platinum is the prevailing one in a PEMFC stack, even though only 0.75 gram, which amounts to 0.012% of the total mass of the stack, is used. In the global warming potential (GWP), the Nafion® material, which is used as the PEM membrane, has the largest environmental impact. The high GWP from the Nafion® material comes from the manufacturing process of the main substances, that is, TFE and perfluoro (alkyl vinyl ether) with sulfonyle acid fluoride, which are part of the perfluorocarbons and fluorocarbons. They have extremely high GWPs (5000-10 000 times that of CO₂), and these extreme values are because they can persist in the atmosphere for thousands of years.72-74 Other materials in the PEMFC stack, except platinum, have a negligible GWP impact compared with Nafion®.

In terms of the human toxicity potential (HTP), the PVdC, which is used as a graphitizeable carbon-forming material for the electrode catalyst in a PEMFC,75,76 has a share of 17.9% in the whole HTP impact indicator, followed by aluminum at 11.2%. Platinum accounts for the largest share of 63.5% in

**TABLE 7** Absolute values of environmental indicators for potential PEMFC stack materials (per 1 gram of material)

| Material                  | ADP elements [kg Sb eq.] | ADP fossil [MJ] | AP [kg SO₂ eq.] | EP [kg Phosphate eq.] | FAETP inf. [kg DCB eq.] | GWP 100 years [kg CO₂ eq.] | HTP inf. [kg DCB eq.] | MAETP inf. [kg DCB eq.] | ODP steady state [kg R11 eq.] | POCP [kg Ethene eq.] | TETP inf. [kg DCB eq.] |
|---------------------------|---------------------------|-----------------|-----------------|-----------------------|--------------------------|---------------------------|----------------------|------------------------|-----------------------------|------------------|------------------------|
| Nafion                    | 1.31E-07                  | 2.12E+00        | 4.10E-04        | 9.14E-05              | 6.38E-06                 | 8.31E-01                 | 3.79E-05            | 2.19E-02               | 1.83E-15                     | 9.31E-06         | 4.30E-07               |
| Glass fibers              | 9.11E-08                  | 2.20E-02        | 1.17E-05        | 6.37E-07              | 1.59E-06                 | 1.73E-03                 | 9.31E-05            | 9.33E-02               | 1.12E-15                     | 6.18E-07         | 1.93E-06               |
| PEEK                      | 2.15E-08                  | 3.42E-01        | 5.23E-05        | 4.22E-05              | 8.28E-05                 | 1.74E-02                 | 6.59E-04            | 9.13E-01               | 4.32E-14                     | 4.25E-06         | 1.23E-05               |
| PTFE Mix                  | 1.60E-06                  | 2.04E-01        | 4.06E-05        | 2.86E-06              | 3.25E-05                 | 1.21E-02                 | 4.36E-04            | 7.87E-01               | 6.06E-10                     | 2.97E-06         | 9.78E-06               |
| Stainless Steel           | 4.58E-07                  | 3.75E-02        | 2.37E-05        | 1.36E-06              | 1.61E-04                 | 3.35E-03                 | 7.54E-02            | 1.44E+00               | 1.09E-15                     | 1.23E-06         | 2.00E-05               |
| Thermoplastic             | 1.27E-08                  | 8.94E-02        | 5.33E-06        | 1.20E-06              | 1.79E-05                 | 4.28E-03                 | 2.72E-04            | 1.69E-01               | 1.63E-15                     | 8.17E-07         | 3.86E-06               |
| Aluminium                 | 4.11E-09                  | 8.95E-02        | 4.01E-05        | 2.32E-06              | 6.00E-05                 | 8.26E-03                 | 4.10E-02            | 19.66                  | 1.00E-14                     | 2.17E-06         | 1.82E-05               |
| Silicone                  | 2.07E-07                  | 8.79E-02        | 1.88E-05        | 1.77E-06              | 3.89E-05                 | 5.85E-03                 | 1.17E-03            | 6.31E-01               | 7.47E-15                     | 1.82E-06         | 9.38E-04               |
| Carbon black              | 5.02E-09                  | 8.50E-02        | 1.15E-05        | 2.95E-06              | 2.12E-04                 | 2.54E-03                 | 8.91E-04            | 6.86E-01               | 1.09E-09                     | 8.53E-07         | 8.53E-06               |
| Graphite                  | 1.17E+00                  | 9.15E-04        | 5.32E-07        | 1.37E-07              | 1.89E-05                 | 7.55E-05                 | 3.68E-05            | 6.84E-02               | 8.35E-12                     | 4.08E-08         | 3.67E-07               |
| Steel product             | 9.39E-09                  | 2.99E-02        | 1.13E-05        | 5.14E-06              | 3.47E-03                 | 2.19E-03                 | 2.84E-03            | 1.45E+01               | 1.11E-10                     | 1.18E-06         | 6.00E-05               |
| Palladium                 | 5.37E-04                  | 5.74E+01        | 1.80E+00        | 3.79E-02              | 1.68E+01                 | 5.12E+00                 | 1.43E+01            | 3.37E+04               | 2.38E-07                     | 7.25E-02         | 2.85E-02               |
| Platinum                  | 2.24E-03                  | 3.11E+00        | 2.53E+00        | 2.69E-01              | 8.53E+00                 | 2.85E+01                 | 9.30E+01            | 2.14E+05               | 8.58E-07                     | 1.04E-01         | 1.39E-01               |
| PVdC                      | 2.62E-08                  | 6.41E-02        | 2.68E-05        | 1.31E-06              | 3.28E-03                 | 4.53E-03                 | 1.79E-02            | 3.47E-01               | n.a.                        | 2.12E-06         | 1.99E-05               |
| Ruthenium                 | 3.71E-04                  | 6.31E-01        | 7.86E-02        | 3.44E-03              | 5.36E-03                 | 6.43E+00                 | 4.54E-01            | 6.24E+02               | 2.45E-10                     | 3.70E-03         | 8.04E-03               |

*Data not available within generic database.

The green-colored cells represent low, the yellow cells represent mid and the red cells represent a high environmental impact in the selected environmental impact indicator of the CML2001 methodology.
the total HTP indicator. In the terrestrial ecotoxicity potential (TETP), the largest impact comes from chromium steel (67%), followed by Pt, which has a 25.8% share in the total TETP indicator. According to the results of the manufacturing phase of the PEMFC stack, with the mass ratios taken from Table 3, Pt and Nafion are the main contributors to the environmental impact of the PEMFC stack. These two materials are also critical according to the presented criticality assessment in Section 3 (Table 2).

In the BoP components of the 1-kWe PEMFC system, chromium steel, aluminum, and HDPE plastics have, on average, the largest environmental impact of all the environmental impact indicators. For the presented PEMFC case study of the manufacturing phase, where precious metals are not included in the LCI of the BoP, it can be concluded that the BoP system represents a minor environmental impact compared with the PEMFC stack (shown in Figure 4).

5.2.2 Environmental impacts of the operating phase

In the operating phase, four scenarios of hydrogen production are analyzed for 20,000 h of operation. The results of the manufacturing phase are compared with four different operating scenarios in Table 11, which shows a comparison of the manufacturing and operating phase results for all the environmental indicators. The operating phase for 20,000 h has a much larger environmental impact compared with the manufacturing phase. Electrolysis powered by hydroelectricity (scenario 2) and NG-SR (scenario 4) is the best scenario with the lowest environmental impact.

In scenario 4 with NG-SR, five out of eleven environmental impact indicators are larger in the case of manufacturing than in the operating phase. But in the case of scenario 2, eight out of eleven environmental impact indicators in the manufacturing phase are larger than in the operating phase. This means that the produced hydrogen could have a lower environmental impact if the hydrogen production technology is carefully selected.

From the analysis of the GWP, it is evident that in all scenarios the GWP of the operating phase is dominant over the manufacturing phase. However, the ODP, as another global environmental indicator, is larger in the manufacturing phase for all the observed scenarios. The regional and local environmental indicators addressing toxicity (TETP, MAETP, and FAETP) are smaller in the case of NG-SR and electrolysis powered by hydroelectricity. This means that the materials used in the PEMFC manufacturing phase have major environmental impacts and these materials have to be additionally and carefully addressed in the EoL phase in order to maximize the recycling potentials of critical materials.

There is a clear difference in the operating phase of the four scenarios, which can be seen from the results in Table 12, where the environmental impact indicators are presented for 1 kWh of generated electricity, including the manufacturing and operating phases. With the results in this form, the effects of the hydrogen-generation scenarios can be easily compared with other FCH technologies. In the last column, the environmental impact indicators for a 5-kW electrical power PEMFC system fueled with methanol, which is produced from natural gas, are presented and compared with the aforementioned scenarios. The results for the 5-kW PEMFC system are taken from the recently concluded H2020 EU project HyTechCycling.28

The hydrogen production method is very important in terms of the environmental impacts when comparing different
**TABLE 9** The shares in CML2001 environmental indicators for materials in 1 kWe PEMFC stack

| Material            | ADP elements, [kg Sb eq.] | ADP fossil, [MJ] | AP, [kg SO₂ eq.] | EP, [kg Phosphate eq.] | FAETP inf., [kg DCB eq.] | GWP 100 years, [kg CO₂ eq.] | HTP inf., [kg DCB eq.] | MAETP inf., [kg DCB eq.] | ODP, steady state, [kg R11 eq.] | POCP, [kg Ethene eq.] | TEETP inf., [kg DCB eq.] |
|---------------------|---------------------------|------------------|------------------|------------------------|--------------------------|----------------------------|----------------------|--------------------------|-----------------------------|---------------------|--------------------------|
| Nafion®             | 0.5%                      | 30.3%            | 1.5%             | 3.0%                   | 0.0%                     | **66.2%**                  | 0.0%                 | 0.0%                     | 0.0%                        | 0.8%                | 0.0%                     |
| Glass fibers        | 0.5%                      | 0.5%             | 0.1%             | 0.0%                   | 0.0%                     | 0.2%                      | 0.0%                 | 0.0%                     | 0.0%                        | 0.1%                | 0.0%                     |
| Aluminum            | 0.1%                      | 5.5%             | 0.6%             | 0.3%                   | 0.0%                     | 2.8%                      | 3.5%                 | 0.0%                     | 0.8%                        | 1.4%                |                          |
| Carbon black        | 0.0%                      | 0.0%             | 0.0%             | 0.0%                   | 0.0%                     | 0.0%                      | 0.0%                 | 0.1%                     | 0.0%                        | 0.0%                | 0.0%                     |
| Graphite            | 0.0%                      | 0.8%             | 0.1%             | 0.3%                   | 0.1%                     | 0.4%                      | 0.2%                 | 0.2%                     | 0.2%                        | 0.4%                |                          |
| Chromium steel      | 0.7%                      | 0.8%             | 0.1%             | 0.4%                   | 2.1%                     | 0.4%                      | 7.2%                 | 1.0%                     | 3.0%                        | 0.2%                | **67.0%**                |
| Platinum            | **96.5%**                 | **47.7%**        | **96.2%**        | **95.2%**              | **92.5%**                | 24.3%                     | **63.5%**            | **95.1%**                | **91.7%**                   | **95.1%**           | **25.8%**                |
| PVdC                | 1.7%                      | 14.4%            | 1.5%             | 0.7%                   | 5.2%                     | 5.7%                      | 17.9%                | 0.2%                     | 0.0%                        | 2.8%                | 5.4%                     |
| Stack, abs-values   | 1.74E-03                  | 489              | 1.98             | 0.212                  | 69.1                     | 87.9                      | 110                  | 168000                   | 7E-07                      | 0.082               | 0.404                    |

The bold value is the highest value in the specific environmental impact CML2001 methodology.

**TABLE 10** Shares in the environmental indicators for materials in the BoP of the 1-kWe PEMFC system

| Material               | ADP elements, [kg Sb eq.] | ADP fossil, [MJ] | AP, [kg SO₂ eq.] | EP, [kg Phosphate eq.] | FAETP inf., [kg DCB eq.] | GWP 100 years, [kg CO₂ eq.] | HTP inf., [kg DCB eq.] | MAETP inf., [kg DCB eq.] | ODP, steady state, [kg R11 eq.] | POCP, [kg Ethene eq.] | TEETP inf., [kg DCB eq.] |
|------------------------|---------------------------|------------------|------------------|------------------------|--------------------------|----------------------------|----------------------|--------------------------|-----------------------------|---------------------|--------------------------|
| Cast iron component    | 0.6%                      | 7.1%             | 3.1%             | 2.8%                   | 0.0%                     | 10.2%                     | 0.3%                 | 1.0%                     | 0.0%                        | 2.5%                | 2.0%                     |
| Steel                  | 0.5%                      | 5.8%             | 5.1%             | 3.3%                   | 0.0%                     | 8.8%                      | 0.1%                 | 0.5%                     | 0.0%                        | 11.7%               | 0.3%                     |
| Aluminum               | 2.2%                      | 25.4%            | **40.5%**        | 13.0%                  | 0.3%                     | **35.8%**                 | 26.1%                | 43.7%                    | 0.0%                        | 24.3%               | 0.4%                     |
| Chromium steel         | **96.3%**                 | 17.1%            | 34.7%            | **71.3%**              | **98.7%**                | 24.3%                     | **73.4%**            | **52.6%**                | **89.9%**                   | 25.8%               | **97.3%**                |
| HDPE                   | 0.3%                      | 38.3%            | 14.3%            | 8.1%                   | 0.8%                     | 17.9%                     | 0.2%                 | 1.9%                     | 8.6%                        | **31.5%**           | 0.0%                     |
| PP                     | 0.0%                      | 6.3%             | 2.3%             | 1.6%                   | 0.1%                     | 3.0%                      | 0.0%                 | 0.3%                     | 1.4%                        | 4.1%                | 0.0%                     |
| BoP, abs-values        | **1.41E-04**              | **264**          | **0.074**        | **0.013**              | **16.4**                 | **17.3**                  | **118**              | **33700**                | **2.54E-07**               | **0.007**           | **3.06**                  |

The bold value is the highest value in the specific environmental impact CML2001 methodology.
The inappropriate inclusion of the hydrogen production method could blur the environmental impacts of the critical materials, which can result in a misinterpretation of the manufacturing phase results for the entire life cycle.

### 5.2.3 Evaluation of the results using the Environmental footprint 2.0 LCIA methodology

To further analyze the environmental impact results, an additional Environmental footprint (EF 2.0) LCIA methodology was used. EF 2.0 includes some environmental impact indicators from CML2001, such as Resource use, mineral, and metals (ADP elements in CML2001) and Resource use, energy carriers (ADP fossil in CML2001), but offers a deeper understanding of, for example, eutrophication, climate change, human health effects, water scarcity, and land use. The environmental indicators for manufacturing and all four operating phase scenarios are presented in Table 13.

The environmental indicators in EF 2.0 are adopted from different LCIA methodologies (ReCiPe, USEtox, Aware, etc.), which means the weighting and normalization factors are different from the CML2001 methodology, and thus, so are the absolute values. Furthermore, from the calculation of the shares for the environmental impact indicators the values for the manufacturing and operating phases show a similar trend. In addition, for the comparisons of some categories, the absolute values are almost the same: Climate Change (fossil) in EF 2.0 fits well with GWP in CML2001, the same goes for...
for Acidification terrestrial and freshwater in EF 2.0 and AP in the CML2001 methodology. For Ozone depletion in EF 2.0 in comparison with ODP and POCP in CML2001, they have different units, but the recalculation of absolute values in shares shows the same trend with both LCIA methodologies. From this analysis, it can be concluded that no matter which LCIA methodology is used for the interpretation of the results, similar trends are shown in the environmental impact contribution of the different phases in the shares for the manufacturing and operating phases of the 1-kW PEMFC system.

5.3 Environmental impacts of the End-of-Life phase

Analyzing the recycling potential of critical materials in a PEMFC is challenging because there are not enough data regarding energy balances and waste flows for modeling the recycling processes in detail. One of the primary goals of this paper is reducing the environmental impacts with proper EoL strategies and the use of secondary critical materials. The results for the EoL phase are discussed in three steps. EoL processes as defined in Table 4: In the present study, Pt recycling was modeled (described in Section 4.2.3) with a 76% recovery ratio (Figure 3). In Table 14, the masses of the secondary materials as a result of the EoL phase of the 1-kW PEMFC system are presented. After the Pt recycling process, 0.57 g of Pt was recovered from 0.75 g of virgin Pt in the MEA. According to the LCA methodology, the mass of the secondary material could avoid the need for virgin material in the manufacturing phase of the same product or used in some other product. Since the EoL phase is directly linked to the manufacturing phase, the operating phase will be excluded in the first step.

The secondary materials produced with different recycling techniques, as well as the energy produced in the energy extraction process, are used as the input flows for the manufacturing phase. In Figure 5, the reduction in the environmental impact indicators (from 100% after the manufacturing stage) of the EoL phase, as defined in Table 4, is presented. EoL focuses on addressing the processes, the secondary materials, and the produced energy in EE (electricity, heat). The results of the environmental impacts calculations for the EoL are negative absolute values (which means lowering the environmental impact of the manufacturing phase), for each impact indicator in the EoL phase. These negative values represent avoided impacts of the EoL phase, which are the sum of the environmental impacts of the used EoL processes, the avoided impacts because of the use of secondary materials, and the avoided energy that is extracted in the EoL processes.
The results presented in Figure 5 show that for all the environmental indicators, apart from the TETP impact indicator (linked with impacts on the local soil), there is a significant reduction (up to 70.7% in AP) in the environmental impact of the manufacturing phase when introducing the EoL phase. The TETP increases because of the use of the EoL process EU-28, the hazardous waste (statistical average) (C-rich, worst-case scenario, incl. landfill) in the EoL model. This process was used as a worst-case scenario for the materials where no data for the EoL phase were available. The EoL phase is the same for all the scenarios, so the TETP increases in all cases (Table 15). From these results, it can be concluded that the EoL phase has the potential for a substantial reduction in the environmental impacts for the manufacturing phase of the 1-kW PEMFC system with the proper EoL phase. With the EoL strategy presented in this study (Table 4), a 37.3% environmental impact reduction, on average, could be achieved for the manufacturing phase.

EoL processes as defined in Table 4 with Pt recycling excluded: Since the focus is on exposing and addressing the critical materials in FCH technologies, it is interesting to show the effect of the EoL phase by neglecting the Pt recycling. In this case, no Pt recycling process is included, and no avoided virgin Pt material is introduced to the manufacturing phase. All the other processes (for other materials) are the same as in the previous case (i) when the Pt is recycled.

The results are presented in the diagram shown in Figure 6. It is clear that there is just a minor reduction in 5 out of 11 of the environmental indicators, while the others even increase. This is due to the fact that the MEA is incinerated and remains landfilled as a worst-case scenario—EU-28: Hazardous waste (statistical average) (C-rich, worst-case scenario, incl. landfill).

The increase in the TETP is even higher than in the case of the MEA recycling. The results in this case show that the reduction in the environmental impacts is negligible in the manufacturing phase when critical materials are not recycled.

EoL phase effect for the entire life cycle of a 1-kW PEMFC system: In this section, the EoL effect on the reduction in the

| Material          | Secondary – REC [kg] | Recovery ratio |
|-------------------|----------------------|----------------|
| Aluminum          | 1.01                 | 96%            |
| Platinum          | 0.00057              | 76%            |
| Steel/Cast iron   | 4.96                 | 87%            |
| PP/HDPE           | 1.47                 | 84%            |

**TABLE 14** Masses of secondary materials from the EoL phase
environmental impact for the entire life cycle (operating phase included) of a 1-kW PEMFC system is analyzed with the EoL strategy presented above (Table 4). The results for the entire life cycle of the 1-kW PEMFC system are shown in Table 15, for all four scenarios with the emphasis on the EoL effect.

The average reduction potential of the EoL phase is presented in the last row of Table 15 for each scenario in relative values. From these results, it is evident that the environmentally sounder operating scenarios (H₂ production method) contribute to a higher environmental impact reduction effect of the EoL phase for the entire life cycle. The results show the highest EoL effect for the environmental impact reduction in the 2nd scenario, followed by the 4th and 3rd. The lowest EoL effect on environmental impact reduction, on average, is achieved in the 1st scenario, where the reduction for the entire life cycle of the 1-kW PEMFC is, on average, 3.3%. The EoL phase reduces the environmental impacts of the manufacturing phase; therefore, the absolute values for the operating phase do not change, but the relative contribution of the manufacturing and operating phases change when the EoL strategy is applied. A more detailed diagram is presented for the 2nd scenario (EU Hydro) in Figure 7, which represents the scenario where the hydrogen production method has the lowest environmental impact. The relative impact of the operating phase in this scenario is the lowest among all the scenarios and the effect of the EoL strategy is more influential for the entire life cycle of the 1-kW PEMFC system. The reference point (100%, Figure 7) is the sum of the environmental impacts of the manufacturing and operating (2nd scenario) phases. These reference columns are divided into two parts: environmental impact share of the operating phase (S2-operation) and the share of the manufacturing phase (S2-manufacturing). As mentioned before, the operating phase is not affected by the EoL, rather it is the relative share of entire life-cycle changes.

The EoL effect on the manufacturing phase is presented in the second columns of each indicator in Figure 7 and shows the reduction in the environmental impacts due to the proposed EoL strategy. The highest EoL effect is seen in the MAETP indicator, where the environmental impact reduction for manufacturing and operating is 60% and the manufacturing phase share drops from 92% to 80%, while the operating phase share increases from 8% to 20% for the entire life cycle of the 1-kW PEMFC system. After introducing the EoL phase (REC, EE, LF), it is evident that the acidification potential (AP) reduction is 52.8% for the entire life cycle. The potential reductions for individual environmental indicators vary from 0.3% for the ODP to 60% for the MAETP, where the major contribution comes from material recycling, especially from the platinum. The only indicator that shows a 37.9% increase is the TETP. The main reason for the TETP increase is the hazardous waste incineration and the landfill process that was modeled as a worst-case scenario for the case of remains after the incineration and materials that cannot be recycled. This increases the environmental impacts on the soil and the Terrestrial ecotoxicity indicators. The results for the entire life cycle of the 1-kW PEMFC system shows that, on average, a 23.7% reduction for all the environmental impact indicators could be achieved.

With modeling the EoL phase, the main goal was to show the potential for lowering the environmental impacts.
in the manufacturing phase and consequently for the entire life cycle of the PEMFC system. In the literature, it is stated that 95% of the platinum could be recovered from the MEA,\textsuperscript{77-79} which is higher than in this study. From this analysis, it can be concluded that in the future an even higher reduction in the environmental impacts with proper EoL could be achieved and the recycling of critical materials is of great importance in the circular economy for PEMFC systems.

6 | CONCLUSIONS

The most relevant and commonly used materials in PEMFCs were identified and classified according to the classification methodology with three criteria: (a) EU criticality methodology, (b) hazardousness, and (c) price. The critical materials were identified and evaluated using the LCA methodology, which highlighted the large environmental impacts of the PGMs and precious metals. The case study for a 1-kWe PEMFC system was carried out from the extraction of raw materials to the end of life (“cradle to grave”). The main conclusions for the manufacturing phase are as follows:

- The environmental impact of the PEMFC stack is prevailing.
- Electricity consumption represents a minor share of the total environmental impacts.
- In PEMFC stack manufacturing, the largest contribution to the environmental impacts comes from platinum (Pt).
- In the BoP, the highest impact comes from chromium steel, aluminum and HDPE.

In the operating phase, electrolysis powered by hydroelectricity and NG-SR has the lowest environmental impact. Since the operating phase has the largest environmental impact, the hydrogen production methodology should be carefully selected. In the EoL phase, recycling was used as the main process, with energy extraction (EE) and landfill (LF) used in some cases:

- With the avoided environmental impacts, a 70.7% reduction in the acidification potential (AP) could be achieved in the manufacturing phase.
- The reduction for all the environmental impact indicators of the manufacturing phase is, on average, 37.3% with the proposed EoL strategy.
- Without the recycling of Pt at the EoL, the reduction in the environmental impacts is negligible.
- For the entire life cycle of a 1-kW PEMFC system, a 23.7% reduction, on average, for all the environmental impact indicators could be achieved.

\begin{figure}[h]
\centering
\includegraphics[width=\textwidth]{figure6.png}
\caption{Effect of EoL phase on the environmental indicators when Pt is not recycled}
\end{figure}
A key challenge in the LCA modeling of FCH technologies is the lack of quality inventory data for the manufacturing phase of the state-of-the-art technologies. Further research will be needed to improve the LCI, not only for PEMFCs, but also for other FCH technologies, such as solid-oxide fuel cells, alkaline, and PEM electrolyzers. In the operating phase, inventory data have to be updated with real working regimes and degradation effects for the core technologies, such as efficiency and power degradation of PEMFC stack. To obtain better results for the LCA, LCI data in the EoL phase have to be upgraded, with the emphasis on the recycling of critical and precious materials.

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