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Environmental assessment of proton exchange membrane fuel cell platinum catalyst recycling

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Abstract

A proton exchange membrane (PEM) fuel cell, an alternative to combustion processes that consume fossil resources, is used to convert energy stored in the form of hydrogen into electricity. The membrane-electrode assembly (MEA), the core of this system, contains platinum, a noble metal, which is a limited resource. This paper presents an environmental assessment of a recycling process for the platinum catalyst contained in the MEA of a PEM fuel cell. During this study, four hydrometallurgical platinum recovery processes from Pt/C particles have been developed at the laboratory scale. The considered process alternatives are composed of the four following steps: leaching, separation, precipitation and filtration. Approximately 76% of the platinum can be recovered as $[\text{NH}_4]_2\text{PtCl}_6$ salt using the most efficient process alternatives. In this case, platinum leaching is carried out with a mixture of H_2O_2 and HCl , followed by liquid/liquid platinum extraction and a precipitation step.

The environmental assessment was performed using the SimaPro 8 tool coupled with the EcoInvent 3.1 database. The environmental impacts were estimated for a 25 cm^2 active area MEA considering the production and end-of-life stages of the MEA life-cycle using the CML-IA baseline V3.02 method. The results show that more than half of the main impacts of the MEA life-cycle can be avoided for four relevant impact categories if platinum is recovered in the end-of-life of the product.

Keywords: PEMFC, LCA, platinum, recycling, hydrometallurgy

Abbreviations

(E) Energy

(Em) Emissions

(CCB) Catalyst coated backing
(CCM) Catalyst coated membrane
(CML) Institute of Environmental Sciences of the Faculty of Science of Leiden University
(FW-ecotox) Fresh water ecotoxicity
(GDL) Gas diffusion layer
(GWP) Global warming potential
(ICE) Internal combustion engine
(LCA) life-cycle assessment
(M-ecotox) Marine aquatic ecotoxicity
(MEA) Membrane-electrode assembly
(ODP) Ozone layer depletion
(PEM) Proton exchange membrane
(PEMFC) Proton exchange membrane fuel cell
(PET) Polyethylene terephthalate
(Pt) Platinum
(PTFE) Polytetrafluoroethylene
(RM) Raw material
(T-ecotox) Terrestrial ecotoxicity
(TFE) Tetrafluoroethylene
(TOPO) Trioctylphosphine oxide

1. Introduction

The current growth in energy demand and increase in resource depletion due to energy consumption have led to the development of new systems for energy production. To meet this demand and reduce the environmental impact resulting from energy production, renewable energy sources based on wind or solar energy have been increasingly used; nevertheless, they cannot guarantee stability in the amount of produced energy, partly because of unstable weather conditions. Electrochemical energy storage, in the form of hydrogen, is a good option to overcome the fluctuations of weather. Proton exchange membrane (PEM) fuel cells could be an alternative energy conversion process, whereby electricity is produced by a reaction between oxygen and hydrogen; the only by-products are heat and water. The redox reactions involved are catalysed by platinum nanoparticles present on the electrodes. The core of a PEM fuel cell is a membrane-electrode assembly in which reactions take place and that contains two catalyst layers, both containing platinum. The current study focuses only on the life-cycle of an MEA.

Currently, the global demand for platinum is constant (approximately 200 t per year (Johnson Matthey, 2013)), but very high compared to the available platinum reserves, which are estimated at approximately 10 kt (the total world resources are estimated to be approximately 40 kt (Labbé and Dupuy, 2014)). Moreover,

only 17% of the used platinum is recycled (see Table 1). In addition, in the scenario of large-scale production of PEM fuel cells, platinum extraction costs would represent between 30 and 40% of the fuel cell manufacturing costs; these values were calculated using data from the literature (James and Spisak, 2012). Taking into account the price of this metal and its limited supply, it is necessary to develop an efficient recycling process regarding the end-of-life stage of MEAs from fuel cells. At the same time, the environmental impacts of such platinum recycling processes have to be compared to the burden of primary platinum production.

Table 1: World platinum market in 2012 (Johnson Matthey, 2013).

Until now, several studies have been performed on assessing the environmental impact of the PEM fuel cell life-cycle. A LCA, taking into account the production and use stages, was carried out by Garraín (2011) to compare PEM fuel cell cars to internal combustion engine vehicles. Two comparative environmental studies between PEM fuel cell and ICE cars (Sørensen and Roskilde, 2004; Hussain et al., 2007), have been performed for three life-cycle stages (production, use and end-of-life stages), but without taking into account platinum recycling. Pehnt (2001, 2003) performed a LCA of a fuel cell system for mobile and stationary applications, considering the production, use and end-of-life stages. In these studies, an average catalyst recycling rate was defined by the author. More recently, Simons and Bauer (2015) studied the LCA of a PEM fuel cell system for road passenger vehicle applications. Their study concerned all life-cycle stages of the fuel cell and was based on data provided by the U.S. Department of Energy. However, data concerning the recycling of used MEAs in the end-of-life of the fuel cell are based on the adaptation of similar hydrometallurgical recycling processes. Thus, none of the already published LCA studies dealing with PEM fuel cells includes real data for the recovery of platinum and other MEA components.

In this context, the objectives of the current study are: (i) to assess the environmental impact of the whole life-cycle of a MEA and (ii) to help decision making for choosing a Pt recycling process from these MEAs based on an assessment of the environmental impact via the life-cycle assessment methodology. Thus, a production scenario that only uses primary platinum (i.e., directly from the extraction plant, including transport for consumption in Europe) is compared to four recycling scenarios, including platinum recovered from MEAs via a hydrometallurgical process (Duclos et al., 2016).

2. PEM fuel cells

A MEA is a stack cell comprising two GDLs, two electrodes (an anode and a cathode) and an ion conducting membrane that is occasionally supported by a polymeric gasket (see Figure 1). When the fuel cell operates, hydrogen is oxidised at the platinum surface of the anode into H^+ and electrons. Electrons and H^+ ions migrate to the cathode, respectively, through an electric circuit and through the membrane. At the cathode surface, oxygen is reduced and reacts with H^+ ions to produce water. The reaction is exothermic, and some heat is co-produced. Electrons moving between the electrodes create an electric current.

MEAs are placed between two bipolar plates and have multiple roles: (i) to supply fuel and oxidant to the reactive sites, (ii) to remove reaction products, (iii) to collect the produced current, (iv) to provide mechanical support for the cells in the stack and (v) to evacuate reaction heat. A stack of several MEAs and bipolar plates placed successively between current collectors and packed with two end-plates, nuts and bolts is called the PEM fuel cell. Additionally, gaskets are used to provide the system with gas-tightness and to adjust the compression of the bipolar plates. The number of MEAs in a stack can vary relative to the desired power. The current study was carried out at the MEA scale and not at the whole fuel cell scale.

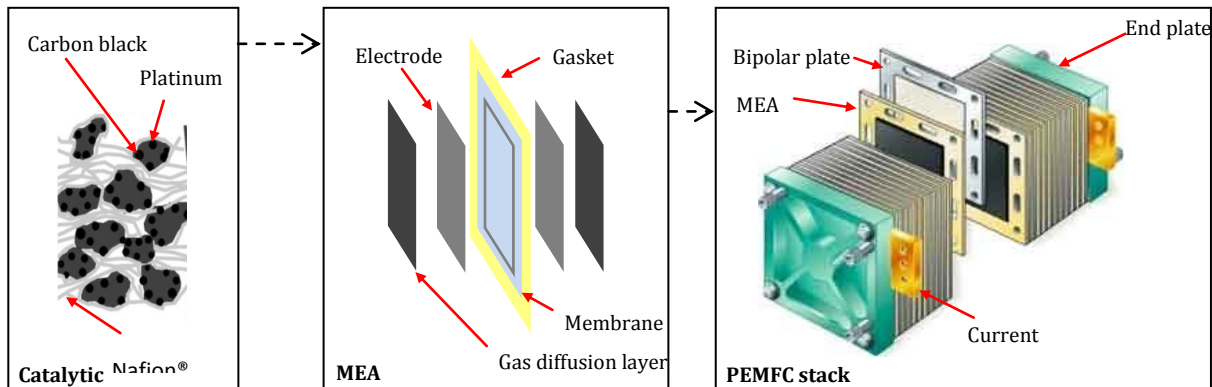


Figure 1: Fuel cell, MEA and electrode components (adapted from: <http://www.theengineer.co.uk>).

2.1. MEA production process

As mentioned in the section above, the MEA is composed of one membrane, two electrodes and two GDLs. The sulfonated fluoropolymer membrane (usually Nafion[®]) and GDL are manufactured separately. The assembly is achieved by applying a so-called “electrode-ink” on one of the components, i.e., either on the membrane named CCM, or on the GDL named CCB. The electrode-ink is obtained by mixing platinised carbon (Pt/C) with a solution containing Nafion[®], water, propanol and other aliphatic alcohols. The actual printing of the ink on the components can be realised at a large scale by two processes: (a) screen-printing (Wang et al., 2015) or (b) inkjet printing (Towne et al., 2007). An additional step can be introduced for CCM manufacturing by forming the catalyst layer on a Teflon support before transferring it onto the membrane by hot pressing (Jung et al., 2012). If the screen-printing process is used, viscous alcohols are added to the ink to obtain a paste. A gasket can be optionally added to the assembly to increase its resistance. Finally, all of the layers are hot-pressed so that solvents evaporate and the layers stick together.

The platinised carbon can be manufactured either by reducing a platinum salt on a carbon powder support by various methods, such as an impregnation reduction method in the liquid or gaseous phase (Veizaga and al., 2012), precipitation reduction method (Liu et al., 2006), or polyols method (Oh et al., 2007) or by sputtering metallic platinum on the carbon support (Fedotov et al., 2013). It has been previously demonstrated that platinum use during this stage has a significant influence on the stack production environmental impact (Pehnt, 2003; Pehnt et al., 2003).

1 Nafion[®], CF₂=CFO-C₃F₆-O-C₂F₄-SO₂F, is produced by copolymerisation of tetrafluoroethylene and a
2 perfluoroalkyl sulfonyl fluoride (Jones, 2012). Then, the membrane is manufactured using an extrusion
3 process at the industrial scale. Finally, the sulfonic groups are hydrolysed by a reaction with a hot solution of
4 NaOH or KOH and the ionomer is converted to the H⁺ form with a strong acid, such as HNO₃.

5 GDLs are made of carbon paper (Park et al., 2008) or carbon cloths (Velayutham, 2011) that are water-
6 proofed using PTFE (between 10 and 20 weight%). The carbon paper is manufactured by hot pressing and
7 carbonizing carbon fibres aggregated with approximately 15% phenolic resin (Hung et al., 2015), whereas
8 carbon cloth is made of knitted carbon fibres. The carbonaceous support is coated with a microporous layer
9 composed of carbon black treated with PTFE to improve PEM fuel cell performance.

15 2.2. Platinum recycling processes

16 A multi-step process had to be carried-out to recycle the platinum from the used fuel cells. The simplest
17 procedures with a negligible environmental impact are mechanical steps, such as the disassembly of a stack
18 into its separate components, i.e., the MEAs, gaskets, bipolar plates, current collectors, and all of the
19 mechanical components (nuts, bolts, etc.). Nevertheless, more complex processes have to be considered to
20 delaminate the different layers constituting the MEA to recover the membrane separately from the catalyst
21 coating, i.e., platinum. Depending on the manufacturing process (see details in § 2.2), the GDL could also be
22 separated mechanically from the CCM. However, mechanical dismantling of the fuel cell was not considered
23 in this study, whereas recycling was only realised at the MEA level with a focus on Pt recovery.

24 Several processes have been developed for platinum recovery from different substrates, such as: (i)
25 pyrometallurgy (metal melting) (Benson et al., 2000), (ii) hydrometallurgy (metal dissolution) (Barakat and
26 Mahmoud, 2004) or (iii) substrate removal by dissolution (Xu et al., 2010) or incineration (Zhao et al.,
27 2014). In the current study, four hydrometallurgical process alternatives are compared to recover platinum.
28 The method developed in this research is efficient and allows high metal purification. Moreover, platinum
29 can be recovered as a solid [NH₄]₂PtCl₆ salt and reused to manufacture new catalyst materials (Verde et al.,
30 2014). The considered process alternatives were developed at the laboratory scale by the authors and are
31 detailed in (Duclos et al., 2016). Each process is thus composed of the following steps: (i) leaching, (ii)
32 extraction (via extraction or resin), (iii) regeneration (of the extractant or the resin), (iv) precipitation, and (v)
33 filtration of the final product i.e., [NH₄]₂PtCl₆ (Figure 2). Two different leachates (acidified solutions of
34 H₂O₂, respectively HNO₃) were used for each recycling process alternative. Moreover, two separation
35 (extraction and regeneration) paths were considered: (1) using ion exchange resin adoption (tested resin:
36 Lewatit-MP-62) and (2) using solvent extraction (tested extractant/diluent couple: Cyanex[®] 923/octanol).
37 The four recycling process alternatives were tested and optimised. To run the experiments, 25 cm² CCM
38 samples with platinum loading of 1 mg·cm⁻² (provided by Paxitech society, Echirrolles, France) were used.
39 These samples were cut into small pieces (4 mm²). The experimental results presented in section §3.3
40 correspond to tests performed on CCMs.

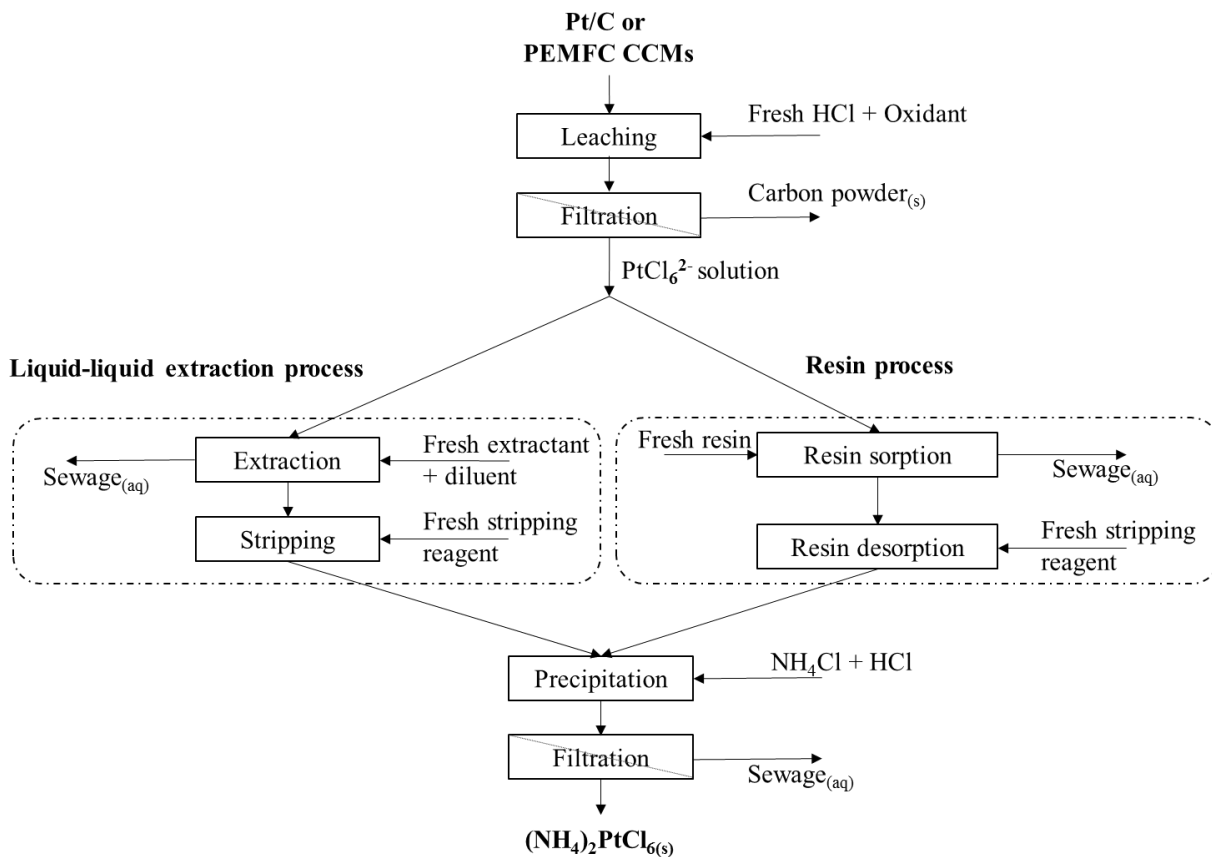


Figure 2: Flow-charts of the two studied platinum recovery processes: (left) the resin recovery process and (right) the liquid/liquid extraction recovery process (Duclos et al., 2016).

3. Environmental impact assessment methodology

LCA is a tool that is used to “assess environmental impacts and potential environmental impacts throughout a product’s life-cycle” from cradle to grave (i.e., from raw material extraction, to disposal and recycling of the final product after use). The ISO 14040 and 14044 standards provide general guidance to perform a LCA, which is composed of four main phases: (1) goal and scope definition, (2) inventory analysis, (3) life-cycle impact assessment and (4) interpretation.

Modelling and simulation of the impacts was performed using the SimaPro 8 tool coupled with the EcoInvent 3.1 database. The environmental impacts were assessed using the CML-IA baseline V3.02 method. This method was used previously by Zucaro et al. (2013) in a LCA of a molten carbonate fuel cell, which is in accordance with the guideline for performing LCAs on fuel cells (Masoni and Zamagni, 2011).

3.1. Step 1: goal and scope definition

The first objective of the current LCA study is to note the main environmental burdens associated with MEA production. The second goal is to identify which of the four platinum recovery process alternatives has the lowest environmental impact and to integrate the chosen alternative in the LCA of a MEA. Thus **scenario**

(1), using primary platinum, is compared to a scenario (2), using primary and recycled platinum (see Figure 3).

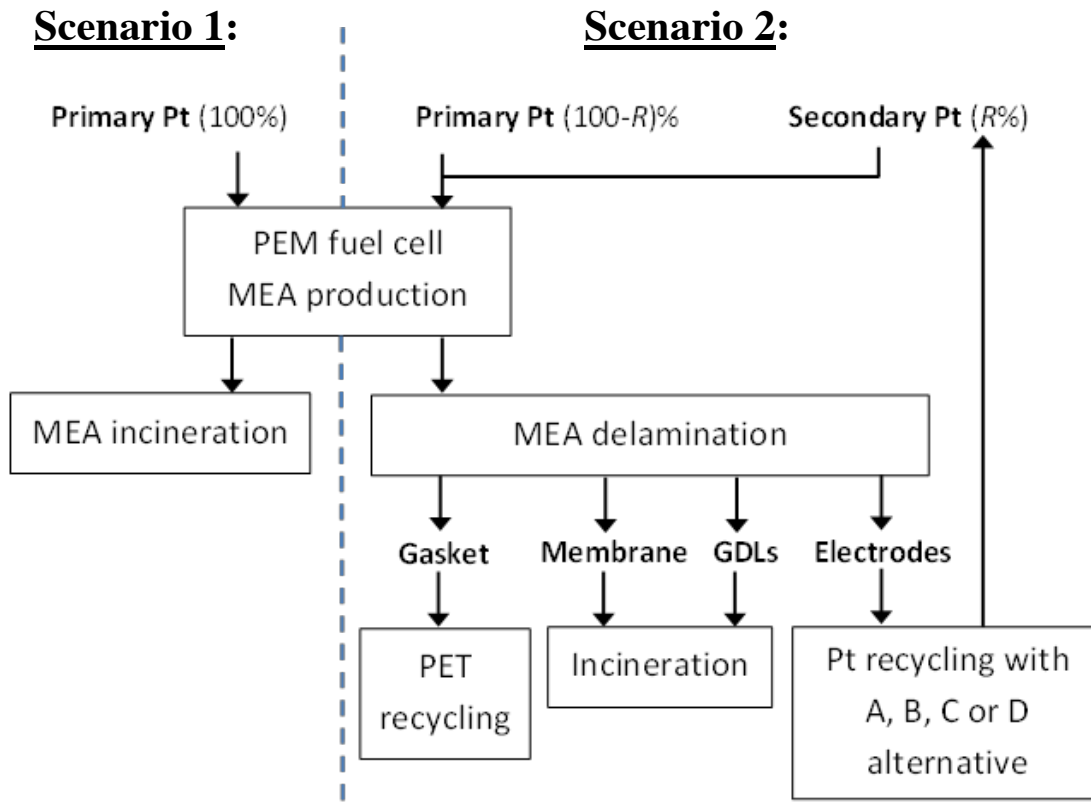


Figure 3: Global model diagram of incineration and recycling scenarios, where R is the recovery efficiency of a process alternative.

The impact is assessed at the MEA scale for technical reasons explained in section §2.3. Thus, the functional unit was defined as: **manufacturing and recycling one MEA that disposes of 25 cm² of active area** (50 cm of electrode layer). Nevertheless, according to the recommendations made by the FC-Hy Guide (Masoni and Zamagni, 2011), the results can further be easily correlated to the electrical power at a stack scale based on the power density (approximately 0.7 W·cm⁻²).

The main stages of the MEA life-cycle are: (i) production stage (i.e., assembly of the membrane, electrodes and gas diffusion layers), (ii) transport to the use facility, (iii) use stage (within a vehicle, in the case of a transport application, for example) and (iv) end-of-life (incineration or platinum recycling). However, only the production and the end-of-life stages are considered here. Neither use stage, which refers to the use of H₂ as fuel, nor the transport stage are taken into account.

Actually, depending on the production process of H₂, e.g., from biomass (gasification or biological production by microorganisms), by solar energy, by wind energy or from fossil fuels, the impacts could be more or less important. Moreover, several studies have already been performed to evaluate the environmental impacts of various production processes of H₂ (Bhandari et al., 2013; Iribarren et al., 2014). It was previously demonstrated (Pehnt, 2001) that the manufacturing stage has a significant influence on the

1 generated environmental impacts over the whole life-cycle of a PEM fuel cell compared to hydrogen
2 production (i.e., use stage). This fact has further been confirmed by two more accurate studies (Pehnt, 2003;
3 Simons and Bauer, 2015), which compared various processes for hydrogen production. However, the life-
4 cycle of H₂ production, i.e., the use stage of PEM fuel cell, does not include any platinum consumption.
5 Once a H₂ production process is fixed, this phase could be considered to be a constant black-box in the life-
6 cycle of a MEA (and PEM fuel cell as the final objective).
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9 Concerning the transport phase, data for the inventory depend on the chosen scenario, i.e., production and
10 use in Europe, or other continents, and so on. Data for this phase are very scarce and not of interest for the
11 studied case. Indeed, the main interest of this study is to compare the environmental impact of the use of
12 primary platinum for PEM fuel cell production, with the use of secondary platinum from a
13 hydrometallurgical recycling process. In the future, integration in the LCA at the PEM fuel cell scale could
14 be envisioned easily.
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19 3.2. Step 2: inventory analysis

20 Data concerning the inventory were mainly taken from the EcoInvent 3.1 database. Nevertheless, several
21 processes and materials had to be defined to build a reliable model (see Table 2). The new processes and
22 materials built are described in detail below.
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28 *Production stage.* Four main compounds are included in the model to simulate the production stage of a
29 MEA: (i) the membrane, made of a sulfonated fluoropolymer; (ii) the GDL, made of PTFE waterproofed
30 carbon black and carbon paper; (iii) the gasket for the membrane, made of polymer whose composition is
31 close to PET; and (iv) the catalyst layers, made of Nafion[®] and carbon supported platinum (Pt/C). The
32 considered composition per FU is given in Table 2, along with references.
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38 **Table 2: MEA manufacturing data.**

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41 Data on energy consumption and raw material consumption for the production stage were obtained from the
42 LITEN department of the CEA, which manufactures MEAs for research applications using the screen
43 printing method. However, for reasons of confidentiality, some information cannot be provided by the
44 manufacturer. The missing data were replaced with data found in the literature (see details in Table 2).
45 Regarding GDL and membrane pre-treatment, material losses of, respectively, 15 and 20% were considered
46 during the MEA manufacturing stage. Concerning catalyst synthesis, liquid phase impregnation reduction
47 and sputtering methods were chosen to model Pt/C production, respectively, from platinum salt or pure
48 platinum metal.
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55 As shown in Table 2, some objects cannot be found in the EcoInvent database; therefore, new materials were
56 defined to simulate, as precisely as possible, the MEA composition (Table 3); whereas some examples are
57 presented below:
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- Nafion[®], which is a sulfonated tetrafluoroethylene based fluoropolymer-copolymer, found in the membrane and the catalyst layers,
- carbon supported platinum (Pt/C) found in the catalyst layers.

Material amounts were calculated according to the composition of new materials. In this case, energy consumption and material losses were considered negligible and 100% synthesis yields were considered. For example, PTFE and sulfuric acid contents, respectively, $\%_{PTFE}$ and $\%_{H_2SO_4}$, were calculated with Eq.1 and Eq. 2. In Eq.1, Nafion[®] is considered to be a product containing SO₃ groups and PTFE. Eq.2 demonstrates the calculation of the amount of sulfuric acid needed to produce the Nafion[®] sulfonic groups.

$$\%_{PTFE} = 1 - \frac{M_{SO_3}}{M_{Nafion}} \quad (1)$$

$$\%_{H_2SO_4} = \frac{M_{SO_3}}{M_{Nafion}} \times \frac{M_{H_2SO_4}}{M_{SO_3}} = 0.15 \times \frac{M_{H_2SO_4}}{M_{SO_3}} \quad (2)$$

where M_x represents the molar mass of chemical x.

Table 3: Additional products in SimaPro.

The EcoInvent database includes several “default” processes, some of them being very precise and specific. All processes are characterised from cradle to grave, including energy and raw material consumption and emissions. Nevertheless, a couple of new processes have been redefined and modelled with SimaPro for the current study:

- evaporation processes of various solvents used during the production stage (e.g., glycerol, propanol and water evaporation), considering chemical emissions to the air
- energy consumption of the MEA assembly process.

End-of-life stage: incineration scenario (1). The end-of-life of the gasket and catalyst layers was modelled with SimaPro using the “Hazardous waste, for incineration” process from EcoInvent database, designed for special waste with an unknown composition. The impact of hydrogen fluoride emissions due to the incineration of products containing high fluorine amounts was modelled by creating new waste treatment processes using the EcoInvent “Waste polyvinylfluoride” incineration process. The burden of the polyvinylfluoride (PVF) incineration process was calculated using Eq.3 as $\%F_{product}$ and $\%F_{PVF}$, which are the fluoride contents of a product and of PVF, respectively. The incineration processes modelled for Nafion[®] and PTFE to assess the membrane and GDL end-of-life impact are shown in Table 4. The GDL i.e., the microporous layer, and carbon paper contain 15% PTFE and 85% of other organic compounds, so its end-of-life was modelled by creating a new waste scenario taking into account 15% of the PTFE treatment process and 85% of the “Hazardous waste, for incineration” treatment process.

$$PVF \text{ incineration} = \frac{\%F_{product}}{\%F_{PVF}} = \frac{\%F_{product}}{0.53} \quad (3)$$

Table 4: Additional waste treatment processes in SimaPro.

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End-of-life stage: recycling scenario (2). In the case of GDL or membrane recycling, the first step in the recycling process is MEA delamination. Nafion[®] (contained in membrane and electrodes) can be dissolved in an autoclave with a water/alcohol mixture under high pressure and temperature (Stephen and Walther, 2005). The membrane can also be separated from MEAs with a water/alcohol mixture at 100 °C (Shore, 2006) or at ambient temperature in an ultrasonication process (Oki et al., 2009). For new MEAs, GDL removal can be performed manually and can also be performed in water by stirring (Oki et al., 2009). As there is no reliable information on the material and energy consumption for this step, it has not been included in the inventory.

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The end-of-life processes of the four components of the MEA were modelled by incineration or recycling, depending on the available data, as follows: (i) the membrane and the GDL are treated by the waste incinerators defined in Table 4 for the previous scenario, (ii) the gasket made of PET is recycled and (iii) the catalyst layers are recycled to recover Pt using the four process alternatives developed within this research project (namely, A, B, C and D).

Data included in the LCA inventory, i.e., electricity consumption (measured with consumption meters), raw material consumption and emissions (to water, air, and waste), are taken from laboratory tests performed in batch mode on Pt recovery from CCMs. The efficiencies of the four recycling processes are given in Table 5, and the complete process inventory used for process modelling on SimaPro is given in Table 6. Every product mass or energy amount is given for 1 kg of used platinum treated with the recycling processes. Thus, the values for one used MEA can be calculated in the same units by multiplying the given amounts by $2.5 \cdot 10^{-5}$. For each reagent available in aqueous solution, the mass given in Table 6 was calculated considering the product as a pure chemical without taking into account the amount of water, according to the EcoInvent 3.1 database principle. The experimental results (Duclos et al., 2016) show that the highest platinum recovery efficiency can be obtained after treatment with process B (H₂O₂/Solvent process); thus, this process alternative will be used as an example in the following parts of the paper. Each recycling process was modelled with SimaPro as a black box, taking into account the energy, material inputs and outputs and waste treatment processes. A detailed input/output mass and energy balance of the H₂O₂/HCl leaching and liquid/liquid extraction process (B) is presented in Figure 4. Platinum is recovered as platinum hexachloroplatinate ([NH₄]₂PtCl₆) as the main product.

Table 5: Efficiency of the Pt recovery processes (Duclos et al., 2016).

Table 6: Recycling processes energy and mass balance including wastes treatment (amounts per kg of Pt at process inlet).

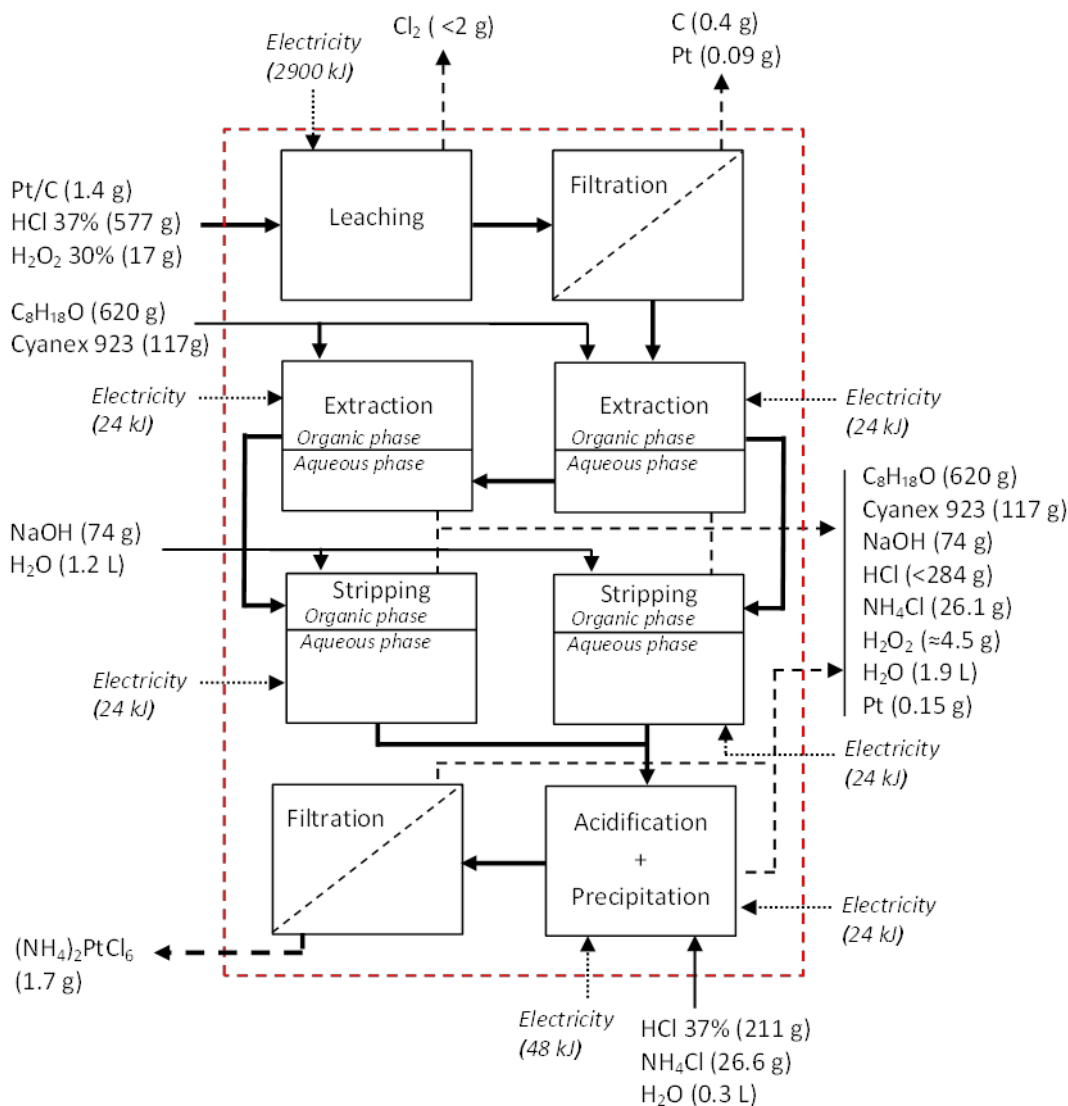


Figure 4: Mass and energy balance of the platinum recovery process from the Pt/C of the H₂O₂/Solvent process (B).

The Cyanex[®] 923 extractant is a mixture of trialkylphosphine oxides that can be considered to be trioctylphosphine oxide. One possible pathway for TOPO synthesis is the reaction between C₈H₁₇MgBr (considered as C₈H₁₈ on SimaPro) and POCl₃ (Kadous et al., 2009). Thus, Cyanex[®] 923 production was modelled on SimaPro with the methodology adopted for Nafion[®] (see composition in Table 3).

The waste-resins used in the recycling processes (C and D) are entered in the “Spent anion exchange resin from potable water production” disposal process, which is available in the EcoInvent database; the organic solvents are treated in the “Spent solvent mixture” specific treatment process available in the EcoInvent database for organic products. Nafion[®] and carbon are incinerated in the “Hazardous waste, for incineration” incinerator. The aqueous effluent treatments are modelled using the “Wastewater, average {CH} treatment of capacity 4.7E10 l/year”. Missing processes, such as acid neutralisation units, were created with SimaPro. The

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Cl₂ produced during the leaching stage was treated by bubbling through a NaOH solution¹. The emitted amount was overestimated to calculate the maximal sodium hydroxide solution volume needed to treat the gaseous emissions. This treatment process was not included in the inventory because of its low environmental impact in comparison with the other hydrometallurgical process steps for the four hydrometallurgical alternatives.

After the recovery of [NH₄]₂PtCl₆ powder, new platinised carbon was produced, and new electrodes were manufactured using this recycled Pt salt. The liquid impregnation method, followed by a liquid phase reduction (Veizaga and al., 2012), was chosen to model Pt/C catalysts remanufacture in scenario (2). Indeed this method is very efficient (higher than 99%) and has a low environmental impact in comparison with the other techniques.

The incineration scenario and the recycling scenario, including process B modelled with SimaPro, are described in Figure 5. Products are represented by square boxes and processes by rounded boxes. EcoInvent products and processes are coloured, whereas the newly created ones are white. Full lines represent elementary flows, i.e., raw material, energy and emissions flows and while dotted lines represent logical connections between various products and processes. The main platinum flow, including recycling processes, is represented with a bold dotted line.

¹ http://www.worldchlorine.org/wp-content/themes/brickthemewp/pdfs/chlorine_safety.pdf

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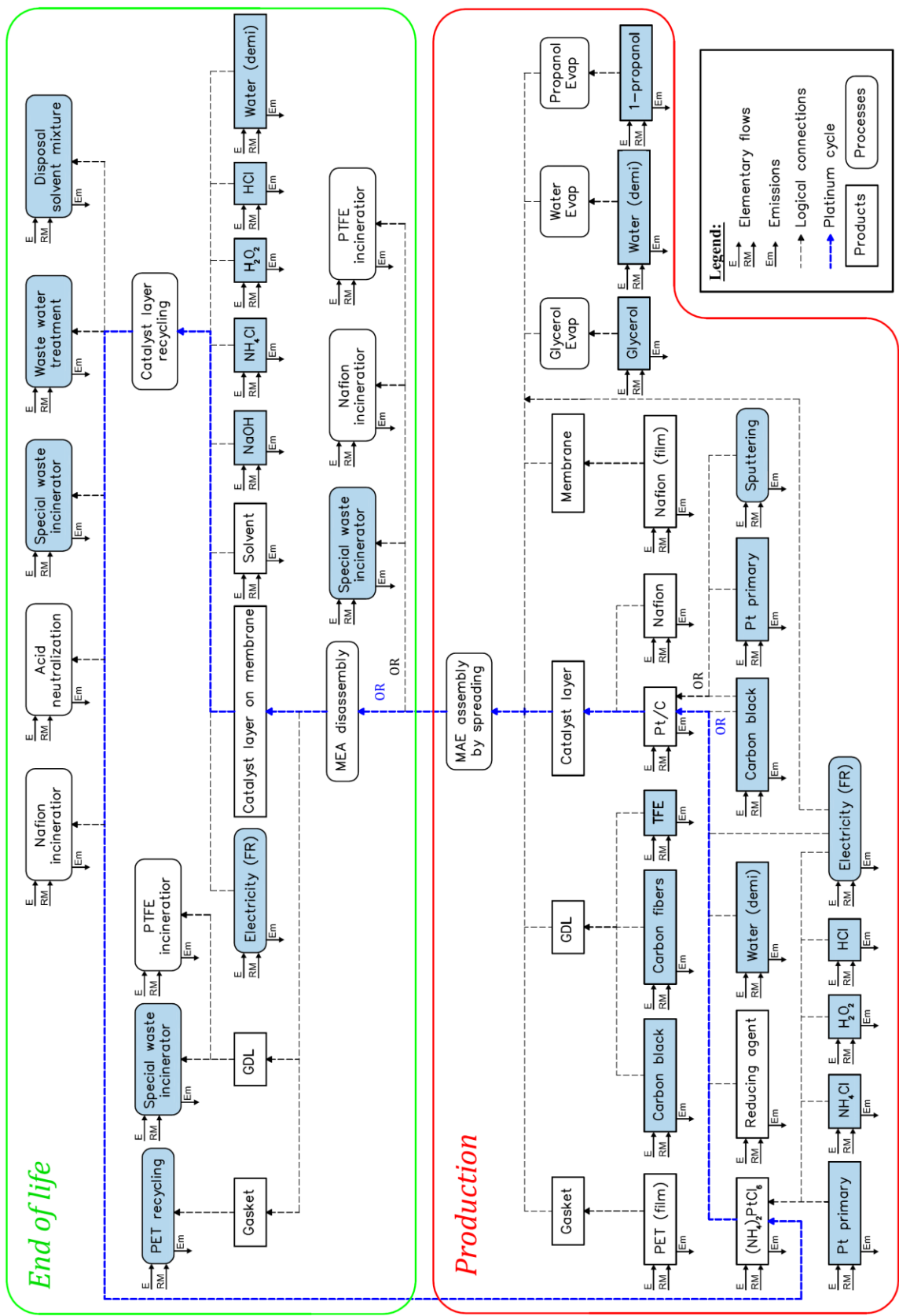


Figure 5: SimaPro model diagram of PEMFC MEA production and end-of-life stages.

3.3. Step 3: life-cycle impact assessment

The impacts are assessed using the CML-IA baseline V3.02 method and the associated impact indicators. Thus, the environmental impact indicators (along with characteristic units) shown in Table 7 are evaluated.

Table 7: CML-IA baseline V3.02 method impact categories.

4. Impact assessment

Within the two considered scenarios, as described in section §3.2, incineration of all components was chosen as the end-of-life option in scenario (1), while recycling was performed in scenario (2). In scenario (1), primary platinum is used for the production stage (i.e., directly from the extraction site) and an open loop scenario is envisaged. For scenario (2), the cycle between production and end-of-life is closed by Pt recycling. Thus, platinum for the production stage is a mixture between recycled (in the end-of-life stage from a used MEA) and primary platinum. The ratio of primary platinum is calculated as a function of the recovery efficiency of platinum in the end-of-life stage. Concerning the end-of-life of a MEA cell, platinum is recovered by four hydrometallurgical process alternatives, as previously described, while other components are incinerated (e.g., membrane, GDL) or recycled (PET support).

4.1. Identification of the most impacting stage for the incineration scenario

The impact was assessed for each process and product of scenario (1), using the CML-IA baseline V3.02 method – the most impacting stage burdens are shown in Figure 6. The results show that platinum production (primary platinum as a raw material) represents the major component of the impacts over the MEA life-cycle for almost all of the impact categories, except the global warming potential and ozone layer depletion. The second main factor is the production of tetrafluoroethylene (mainly used in the membrane manufacturing step), which contributes to each impact and represents, respectively, 40% and more than 99% of the total burdens for the GWP and the ODP categories. Indeed, fluorocarbon gases are emitted during TFE manufacturing; these pollutants have a large global warming potential and lead to ozone layer depletion.

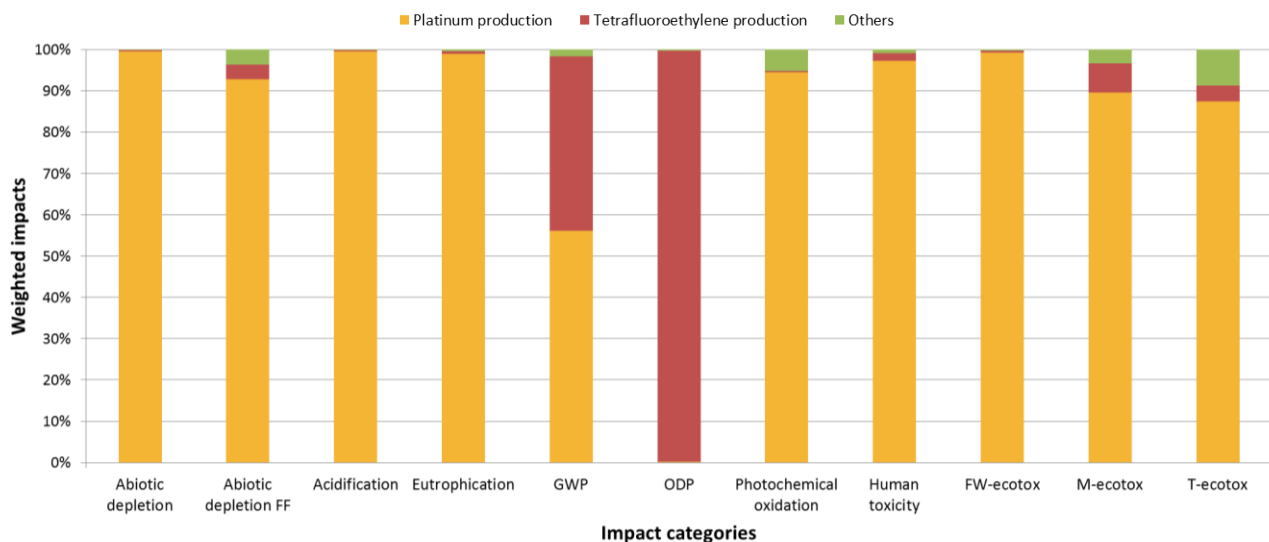


Figure 6: MEA life-cycle hot-spot identification from the scenario (1).

1 Platinum extraction and purification have a large environmental impact mainly because of trace metal
2 (beryllium, mercury, nickel, selenium, etc.) emissions to water and nitrogen oxide, hydrogen fluoride and
3 sulphur dioxide emission to air during pyrometallurgical and hydrometallurgical purification steps
4 (EcoInvent inventory). The results were calculated for CCMs manufactured by Paxitech with a platinum
5 loading of $1 \text{ mg} \cdot \text{cm}^{-2}$. Currently, lower platinum loadings are being used in fuel cells (between 0.1 and 1
6 $\text{mg} \cdot \text{cm}^{-2}$). The simulations performed with such loadings show that platinum use would still represent a large
7 portion of PEM fuel cell life-cycle impacts. Consequently, platinum recovery could be the solution for both
8 reducing the environmental impacts of the life-cycle of MEA (production & end-of-life) and PEM fuel cell
9 cost in the long term in the case of large scale production. Thus, catalyst recovery has to be taken into
10 account before other components' recycling.

11 **4.2. Comparison of the four platinum recycling processes: scenario (2)**

12 The four life-cycle scenarios considering platinum recycling were compared. The results for all of the impact
13 categories were weighted according to the CML-IA baseline V3.02 method to convert the impact units into
14 points; the weighting of each category in the final score is presented in Table 8 for the B process alternative.
15 Three main impact categories can be highlighted from Table 8, i.e., acidification, fresh water aquatic
16 ecotoxicity and marine aquatic ecotoxicity. Concerning the three other processes' recycling scenarios, score
17 weighting shows that these are also the most impacted processes. In addition, abiotic depletion was also
18 selected to take into account metal depletions due to platinum use. Finally, the global warming potential and
19 ozone layer depletion categories were chosen to consider the fluorocarbon gas emissions caused by Nafion[®]
20 manufacture. The results for the aforementioned impact categories are compared in Figure 7 for the four
21 platinum recovery processes. The least impacting recovery process is the B process, while the most
22 impacting is the A process. Indeed, for the three most impacted categories, i.e., acidification, FW-ecotox and
23 M-ecotox, the impact scores of the MEA life-cycle are the lowest in the case of platinum recycling with the
24 B process and the highest with the A process. The B process is more impacting than the C and D processes
25 for GWP. Furthermore, the C and D processes have the same potential environmental burdens for all of the
26 impact categories investigated.

Table 8: Impact categories burdens in the life-cycle normalised impact score for the scenario (2)-B process.

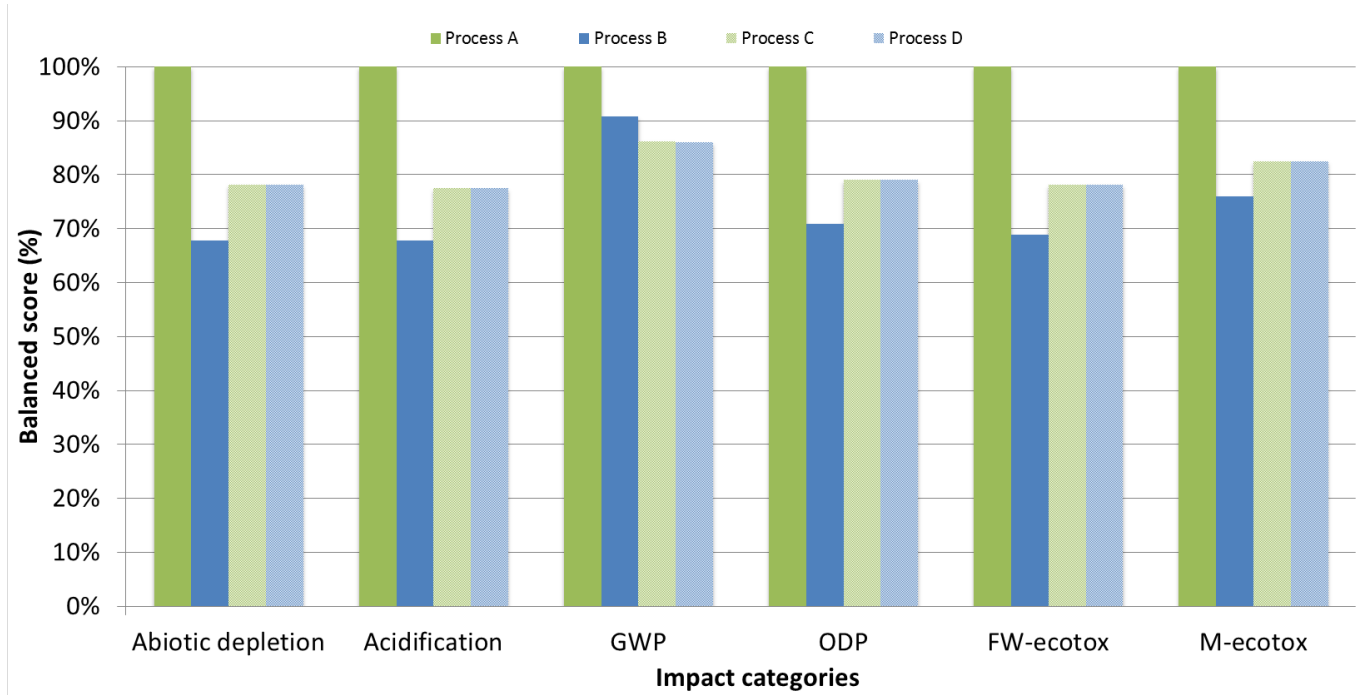


Figure 7: Impact assessment of a 25 cm² PEM fuel cell MEA considering production and end-of-life stages with scenario (2).

Integrated in the whole life-cycle of the MEA, the impact ratio mostly depends on the recovery efficiency of platinum (Table 5); the more platinum is recovered from the used MEA, the more is used in the production stage. Thus, less primary platinum is consumed. As shown in scenario (1) (see Figure 6), primary platinum is the most impacting of all of the components used in the manufacture of the MEA.

4.3. Comparison of the recycling and incineration scenarios

The best process, in terms of potential impact, for the recovery of platinum from used MEAs, i.e., the B process (H₂O₂/Solvent), was integrated into the whole life-cycle of the MEA and compared to the scenario of incineration (1). The environmental impact was assessed for each production process and product; the results are shown in Figure 8. Platinum catalyst recycling with the H₂O₂/Solvent process results in a decrease of more than 60% in the main impact category burdens (Table 8: acidification, abiotic depletion, FW-ecotox and M-ecotox) and more than 30% of the GWP category over the whole life-cycle. The ODP category is not impacted by platinum recycling because this category is mostly affected by Nafion[®] production. The results show that primary platinum production remains the most impacting stage, even when MEA recycling is considered. In addition, Nafion[®] production also has a significant influence on the environmental impact of the MEA's life-cycle.

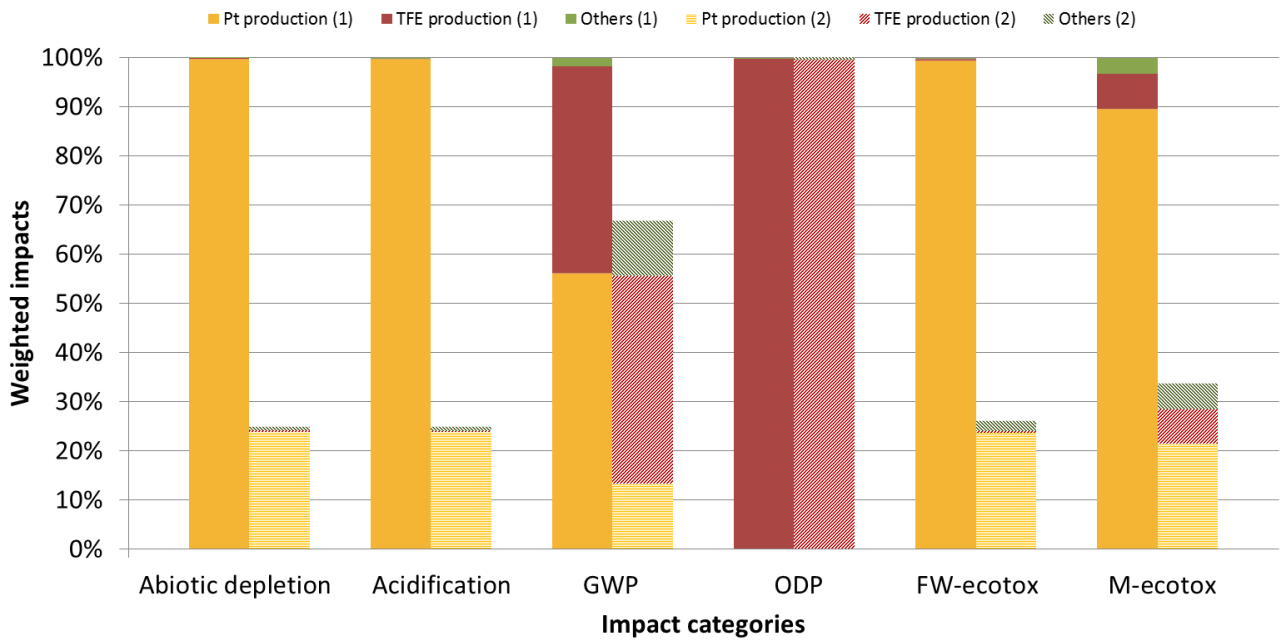


Figure 8: Comparison between the MEA incineration scenario (1) and platinum recycling scenario (2) considering platinum recycling with the H₂O₂/Solvent process.

Despite the fact that the results are in favour of platinum recovery, several aspects need to be considered. Scale-up from the laboratory scale to the industrial scales can radically change these results. Indeed, experiments concerning the current study were performed at the laboratory scale, which implies that energy used for material transport was not evaluated. Moreover, process energy consumption was considered for small sized equipment units, but would differ at the industrial scale. In addition, the MEA disassembly process has not been taken into account in the LCA. Furthermore, tetrafluoroethylene production is the second burden of MEA life-cycle, and its importance in the PEM fuel cell life-cycle is increased, so Nafion[®] membrane recycling should be investigated.

5. Conclusions and perspectives

This study performed the first complete LCA study at the MEA scale. Laboratory data combined with literature data based on valid hypotheses were used to simulate the environmental impacts of the production and end-of-life stages of a 25 cm active area MEA, whereas four different hydrometallurgical processes for platinum recovery were compared in this study.

LCA simulations performed with SimaPro show that the B process alternative (H₂O₂/Solvent recycling process) is the least impacting of the four hydrometallurgical recycling alternatives proposed. Moreover, the assessment reveals that the MEA life-cycle impact can be reduced by 60% if electrode recycling is carried out at the end-of-life stage of the fuel cell by the H₂O₂/Solvent recycling process. In addition, the main impact category decrease is proportional to the platinum recycling rate.

1 The LCA model is based on various simplifying hypotheses and could be improved with a more detailed
2 study. MEA catalyst recycling could be scaled-up at the pilot scale to include a continuous mode (the current
3 study has been performed in batch mode) and the recycling of various solvents (e.g., recirculation of acid,
4 oxidant, etc.). Moreover, a deeper study on the optimisation of the parameters of the recovery process would
5 increase the platinum recovery efficiency and reduce the overall impact.
6

7 Concerning the environmental impact assessment, the inventory could eventually be completed by using data
8 on MEA disassembly and membrane and gas diffusion layer recovery. No attempt was made in this direction
9 in this study; however, some ideas to be considered were explored (see details in § 3.3). This would represent
10 a step forward in membrane and GDL recycling.
11

12 Assuming that the suggested process alternatives for the recycling of MEA would be implemented at the
13 industrial scale, the transport phase to recovery and eventually to the production site should be included in
14 the LCA. On the other hand, the use stage is fully connected to H₂ consumption over the use stage of the
15 PEM fuel cell. Finally, a complete assessment at the MEA scale could be integrated in what would be a first
16 complete LCA study on PEM fuel cells, i.e., including the transport and use stages of the fuel cell.
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Figure 1: Fuel cell, MEA and electrodes components (adapted from: <http://www.theengineer.co.uk>).

Figure 2: Flow-sheets of the two studied platinum recovery processes: (left) resin recovery process and (right) liquid/liquid extraction recovery process (Duclos et al., 2016).

Figure 3: Global model diagram of incineration and recycling scenarios, where R is the recovery efficiency of a process alternative.

Figure 4: Mass and energy balance of platinum recovery process from Pt/C of the H_2O_2 /Solvent process (B).

Figure 5: SimaPro model diagram of PEMFC MEA production and end-of-life stages (platinum recycling with process B).

Figure 6: MEA life cycle hot-spots identification from the scenario (1).

Figure 7: Impact assessment of a 25 cm^2 PEM fuel cell MEA considering production and end-of-life stages with the scenario (2).

Figure 8: Comparison between the MEA incineration scenario (1), and the platinum recycling scenario (2) considering platinum recycling with H_2O_2 /Solvent process.

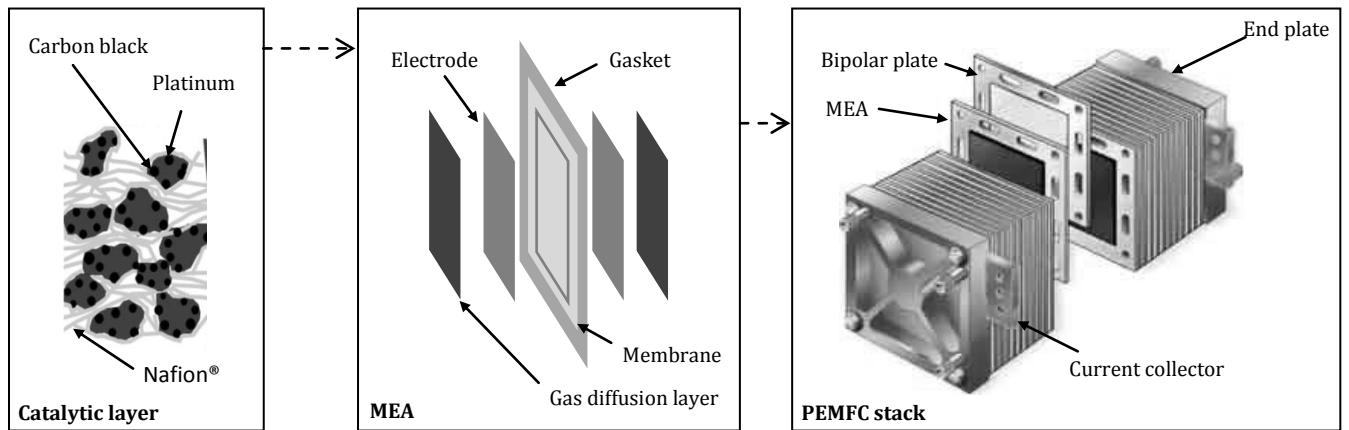


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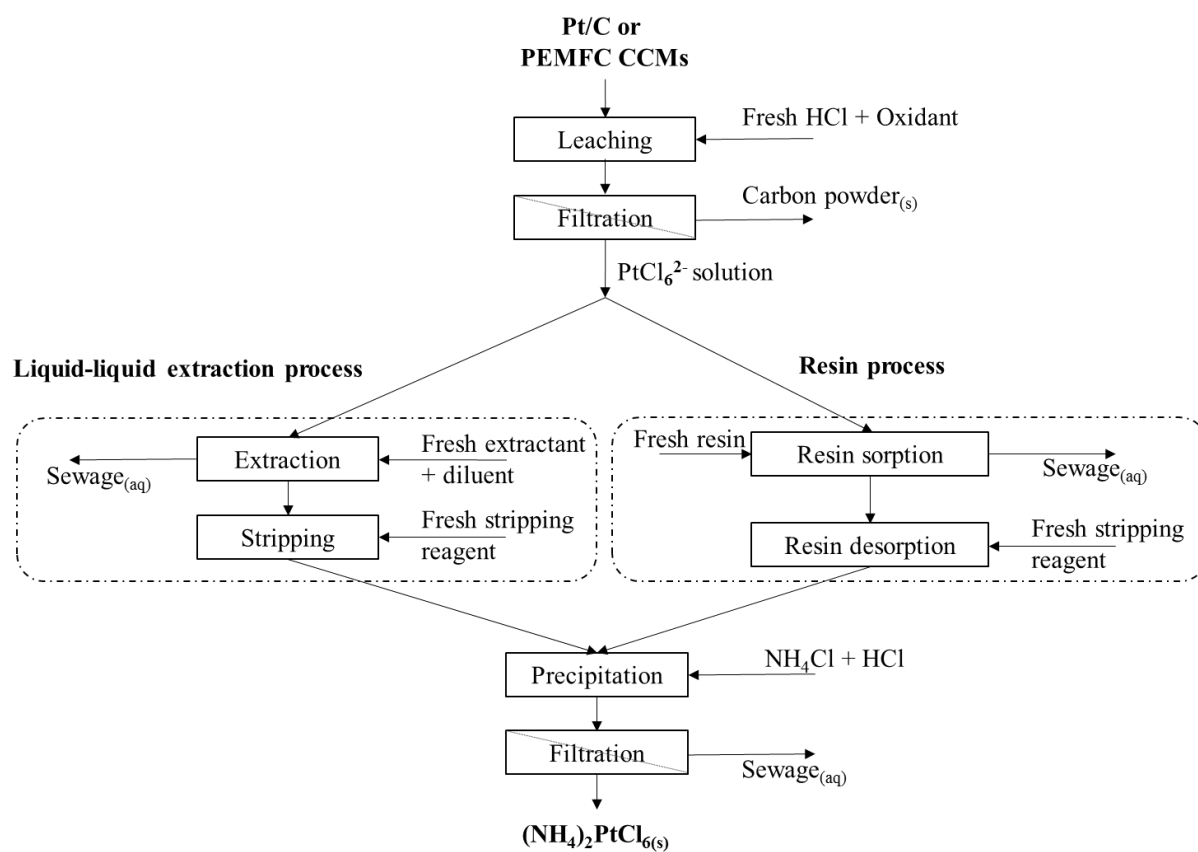


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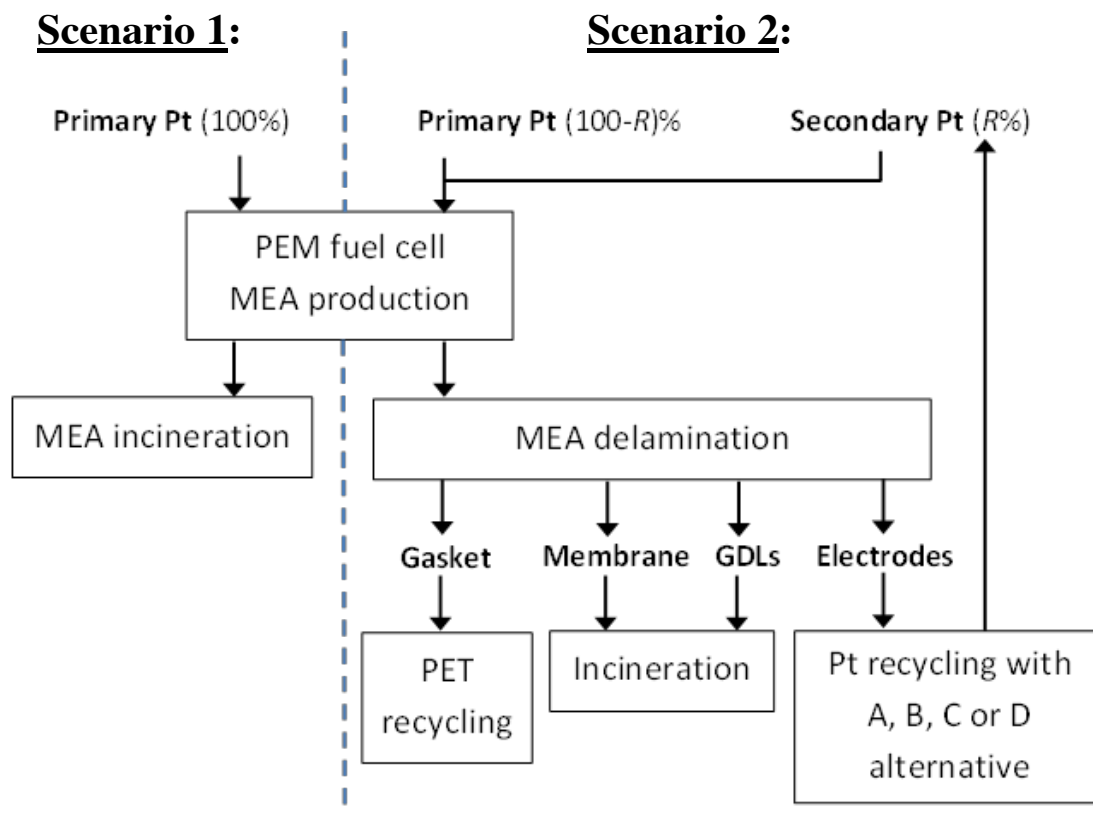


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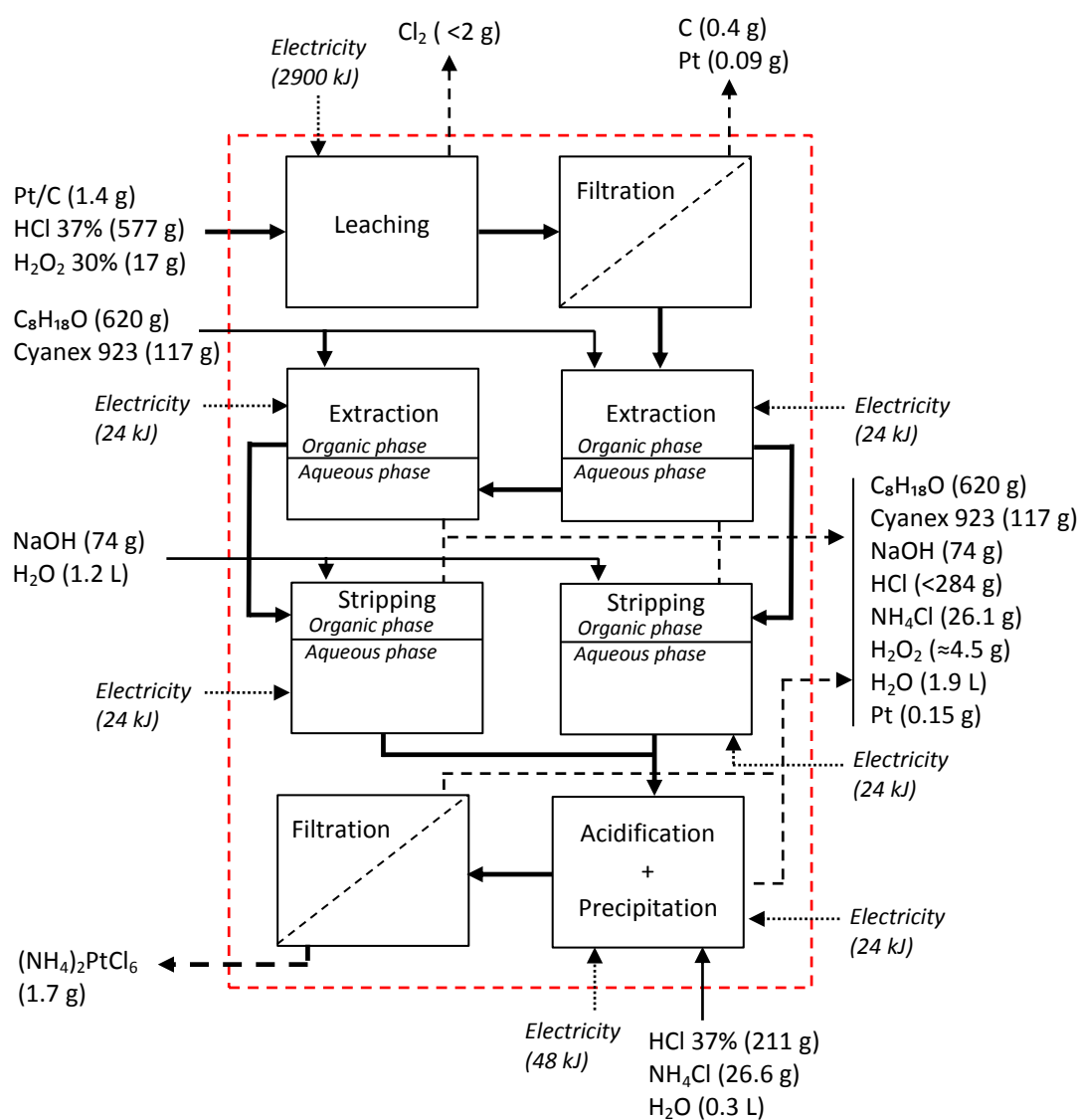


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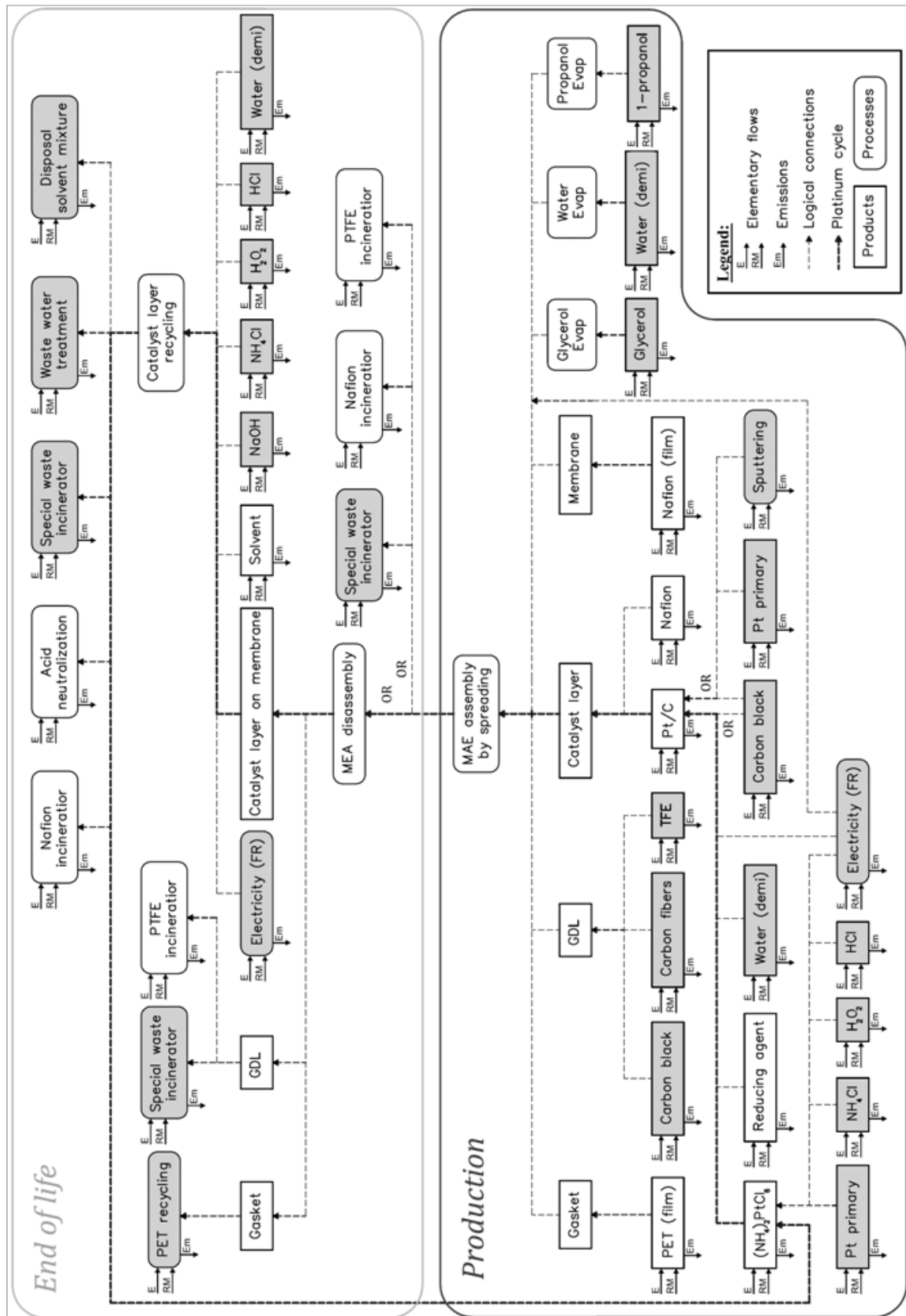


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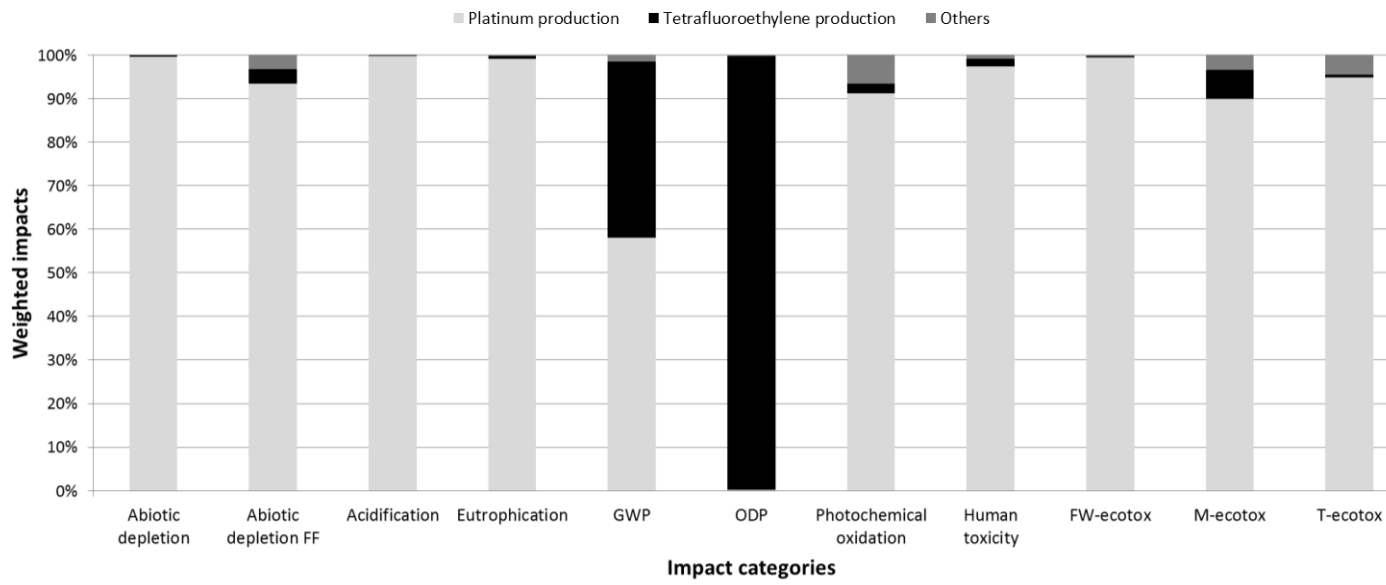


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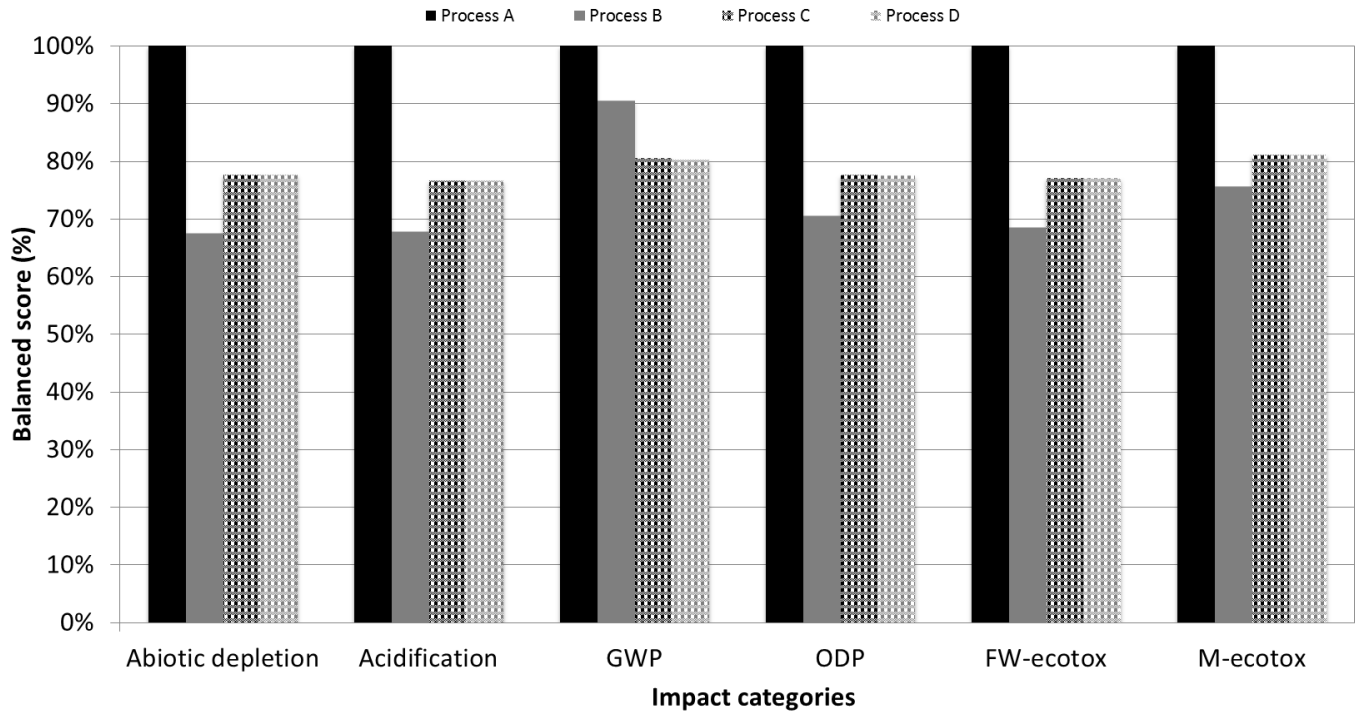


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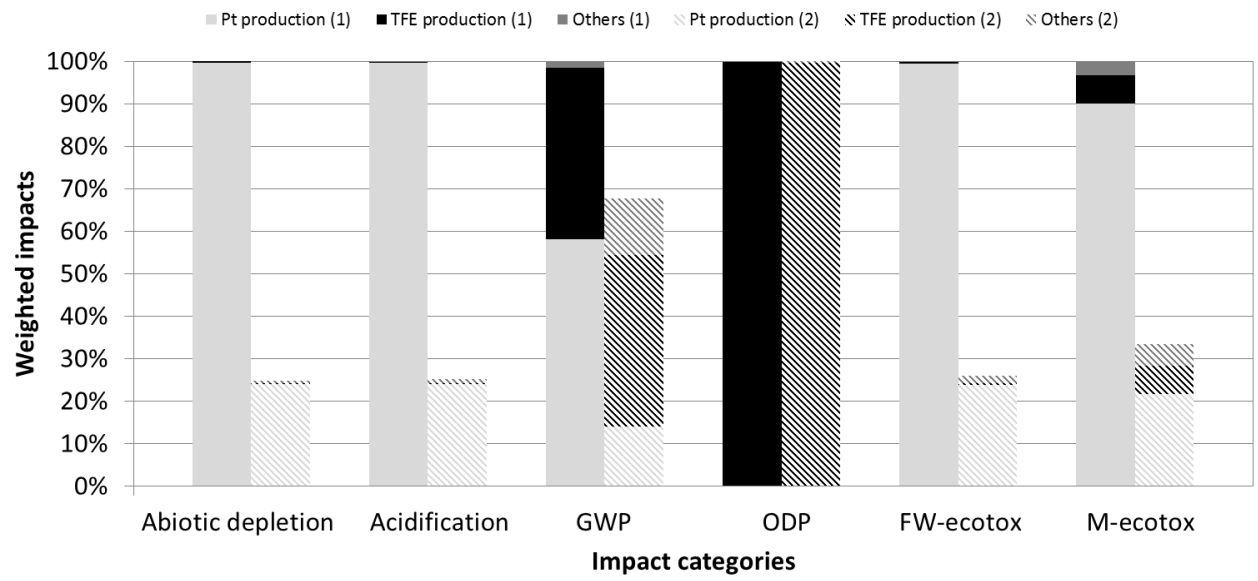


Figure 8: Comparison between the MEA incineration scenario (1), and the platinum recycling scenario (2) considering platinum recycling with H₂O₂/Solvent process.

Table 1: World platinum market in 2012 (Johnson Matthey, 2013).

	Auto-catalyst	Jewellery	Industrial	Investment	Total
Global demand (t)	91	78	44	13	225
Recycling (t)	32	5	0	/	37
Recycling rate (%)	35%	7%	1%	/	17%

Table 2: MEA manufacturing data.

MEA components	Materials	Material/process available in EcoInvent	Mass/MEA surface (kg·m ⁻²)	Reference / information provider
Catalyst layer (ink)	Platinized carbon black	No	1.4·10 ⁻²	Paxitech and (Wang et al., 2015)
	Water	Yes	3.0·10 ⁻²	
	Propanol	Yes	1.1·10 ⁻¹	
	Nafion [®]	No	7.4·10 ⁻³	
	Cyclohexanol	Yes	5.5·10 ⁻²	
	Ethylene glycol	Yes	4.8·10 ⁻²	
Membrane ^a	Nafion [®]	No	5.1·10 ⁻¹	Chemours website ³
GDL (carbon paper) ^a	Carbon fibre	Yes	1.8·10 ⁻¹	(Park et al., 2008) and (Hung et al., 2015)
	Phenolic resin	Yes	3.2·10 ⁻²	
	Carbon black	Yes	4.0·10 ⁻²	
	PTFE	Yes	3.4·10 ⁻²	
Gasket	Approximated as PET	Yes	1.7·10 ⁻²	CEA/LITEN
Manufacturing step	Energy		Energy/MEA surface (MJ·m ⁻²)	Reference
Ink production	Electricity	Yes	5.0	CEA/LITEN
MEA assembly	Electricity	Yes	8.8	CEA/LITEN

^aMembrane and GDL data without to pre-treatment material losses consideration.

Table 3: Additional products in SimaPro.

Material	SimaPro associated product	Composition product/material mass ratios (kg·kg ⁻¹)	Process
Nafion [®]	PTFE + H ₂ SO ₄	0.85 + 0.18	/
Pt/C (70%Pt)	Pt + Carbon black	0.7 + 0.3	Metal sputtering
Cyanex [®] 923	POCl ₃ + C ₈ H ₁₈	0.4 + 0.88	/

Table 4: Additional waste treatment processes in SimaPro.

	Nafion [®] incineration	PTFE incineration
Fluorine content	59%	76%
PVF incineration (kg·kg ⁻¹)	1.1	1.4

Table 5: Efficiency of Pt recovery processes (Duclos et al., 2016).

Recovery process denomination	A	B	C	D
Extraction process	Solvent		Resin	
Leachate	HNO ₃	H ₂ O ₂	HNO ₃	H ₂ O ₂
Leaching	93%	91%	93%	91%
Separation	81%	86%	86%	89%
Precipitation	85%	97%	90%	89%
Efficiency (% of Pt recovered)	64%	76%	72%	72%

Table 6: Recycling processes energy and mass balance including wastes treatment (amounts per kg of Pt at process inlet).

		Process inlets				
	Products/	energy	Process			
			A	B	C	D
Recycling process						
	Nafion® (kg)		0.7	0.7	0.7	0.7
	Pt/C (kg)		1.4	1.4	1.4	1.4
	HCl without water (kg)		279	284	265	269
	HNO ₃ without water (kg)		25	/	25	/
	H ₂ O ₂ without water (kg)		/	5	/	5
	Deionised water (kg)		1900	1900	1700	1700
	Cyanex® 923 (kg)		117	117	/	/
	Pentanol (kg)		620	620	/	/
	Anionic resin (kg)		/	/	40	40
	NaOH without water (kg)		74	74	40	40
	NH ₄ Cl (kg)		26.6	26.6	21.4	21.4
	<i>Electricity low voltage {FR} (MJ)</i>		3000	3000	4000	4000
		Process outlets				
	Wastes or products/ treatment process		Process			
			A	B	C	D
Solid wastes	Pt (kg)		0.36	0.24	0.28	0.28
	C (kg)		0.4	0.4	0.4	0.4
	<i>Hazardous waste, for incineration {GLO} (kg)</i>		0.8	0.7	0.7	0.7
	Nafion® (kg)		0.7	0.7	0.7	0.7
	<i>Nafion incinerator (kg)</i>		0.7	0.7	0.7	0.7
	Anionic resin (kg)		/	/	40	40
	<i>Spent anion exchange resin from potable water production {GLO} (kg)</i>		/	/	80	80
Aqueous wastes	HCl (kg)		279	283	264	268
	HNO ₃ (kg)		24	/	24	/
	<i>Hydrochloric acid neutralization (kg)</i>		279	284	265	269
	<i>Nitric acid neutralization (kg)</i>		25	/	25	/
	Deionised water (kg)		1900	1900	1700	1700
	H ₂ O ₂ (kg)		/	4	/	4
	NaOH (kg)		74	74	40	40
	NH ₄ Cl (kg)		26.2	26.1	21.0	21.0
	<i>Wastewater, average {CH} treatment of capacity 4.7E10l/year (m³)</i>		1.9	1.9	1.7	1.7
Organic wastes	Cyanex® 923 (kg)		117	117	/	/
	Pentanol (kg)		620	620	/	/
	<i>Spent solvent mixture {GLO} (kg)</i>		737	737	/	/
Products	(NH ₄) ₂ PtCl ₆ (kg)		1.47	1.73	1.64	1.64

Table 7: CML-IA baseline V3.02 method impact categories.

Impact category	Unit
Abiotic depletion	(kg Sb eq)
Abiotic depletion fossil fuels	(MJ)
Acidification	(kg SO ₂ eq)
Eutrophication	(kg PO ₄ ³⁻ eq)
Global warming potential	(kg CO ₂ eq)
Ozone layer depletion	(kg CFC-11 eq)
Photochemical oxidation	(kg C ₂ H ₄)
Human toxicity	(kg 1,4-DB eq)
Fresh water aquatic ecotoxicity	(kg 1,4-DB eq)
Marine aquatic ecotoxicity	(kg 1,4-DB eq)
Terrestrial ecotoxicity	(kg 1,4-DB eq)

Table 8: Impact categories burdens in the life cycle normalised impact score for scenario (2)-B process.

Impact category	Category score weighting
Abiotic depletion	0.9%
Abiotic depletion (fossil fuels)	0.5%
Acidification	4%
Eutrophication	0.7%
GWP	0.8%
ODP	0.8%
Photochemical oxidation	0.6%
Human toxicity	0.4%
FW-ecotox	6%
M-ecotox	86%
T-ecotox	<0.1%

*Most impacted categories are marked in bold; less impacting ones in grey