New technologies and strategies for fuel cells and hydrogen technologies in the phase of recycling and dismantling

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Executive Summary

This report constitutes the Deliverable 2.2 on existing end-of-life (EoL) technologies applicable to fuel cells and hydrogen (FCH) products, which is associated with Task 2.2 "Mapping of existing recycling technologies and matching with FCH technologies" within Work Package 2 "Regulatory analysis, critical materials and components identification and mapping of recycling technologies" of the HyTechCycling project. EoL strategies for key FCH products (viz., proton exchange membrane fuel cells/water electrolysers, alkaline water electrolysers, and solid oxide fuel cells) are found to be currently focused on the (hydrometallurgical and pyrometallurgical) recovery of precious metals employed in the stacks as catalysts. Regarding balance-of-plant (BoP) components, the further recovery of valuable materials is considered, e.g. metals and polymers for which mature recycling technologies are available. In order to face effectively the challenge of cost-competitiveness for a well-established hydrogen economy, a full EoL strategy to reduce the costs of FCH devices is still required. The HyTechCycling project goes forward in this direction.

Contents

List of Figures

List of Tables

Abbreviations

AWE	Alkaline water electrolyser
BoP	Balance of plant
CCM	Catalyst-coated membrane
CCB	Catalyst-coated backing
EoL	End of life
FCH	Fuel cell and hydrogen
GDL	Gas diffusion layer
MEA	Membrane electrode assembly
PCB	Printed circuit board
PEM	Proton exchange membrane
PEMFC	Proton exchange membrane fuel cell
PEMWE	Proton exchange membrane water electrolyser
PGM	Platinum-group metals
PTFE	Polytetrafluoroethylene
SOFC	Solid oxide fuel cell
WEEE	Waste electrical and electronic equipment

1. Introduction

The Paris Agreement [1] marks a turning point within the current energy transition context, bringing all nations into a common cause that addresses the global issue of climate change. This agreement requires all parties to implement ambitious efforts to mitigate or adapt to the effects of climate change. In this sense, the European Union has recently launched a Climate Action Roadmap with a target to reduce greenhouse gas emissions by 80% by 2050 with respect to 1990 [2]. To reach this goal, a continued progress towards a low-carbon economy and society is required, with clean technologies playing a crucial role. In particular, the replacement of fossil-based technologies with alternative ones, such as fuel cells and hydrogen (FCH) technologies, can be considered a key pathway to reduce significantly the dependence on fossil sources and the related emissions [3]. Hydrogen arises as a promising alternative with the potential to provide low-carbon energy for all sectors of the economy (e.g., transport, building and industry sectors). However, in order to pave the way for a wide deployment of commercial FCH technologies, several barriers remain to be overcome. In this regard, current hydrogen infrastructure (especially in terms of distribution and use) is insufficient and its suitable development represents a major challenge for a well-established hydrogen economy. Besides, public support at the global and regional level is considered a key need for the success of FCH systems as clean energy technologies in a future low-carbon scenario.

Apart from infrastructure and societal barriers, a wide deployment of commercial FCH technologies is also conditioned by the current lack of well-defined end-of-life (EoL) strategies. Within this context, the HyTechCycling project aims to deliver reference documentation and studies on both conventional and novel EoL technologies and strategies applicable to FCH technologies, paving the way for future demonstration actions and advances in roadmaps and regulations. A key starting point in this project is the thorough study of the current situation regarding the disposal phase applied to FCH technologies, thereby building a sound basis for the remaining activities of the project. Hence, the present report addresses the scanning of existing EoL technologies that are or have the potential to be applied to FCH technologies.

2. Structure of the work

An extensive FCH-oriented literature review on EoL technologies is carried out in this report. From the literature survey, the main components and materials of the target FCH technologies are identified, making a distinction between stack components and balance-of-plant (BoP) components. The HyTechCycling target technologies include proton exchange membrane fuel cells (PEMFC), proton exchange membrane water electrolysers (PEMWE), alkaline water electrolysers (AWE), and solid oxide fuel cells (SOFC).

After the identification of key components and materials, specific EoL strategies available for the assessed FCH technologies are identified. Furthermore, conventional/existing recycling technologies applicable to relevant materials of the different components are documented.

3. Description and components of Solid Oxide Fuel Cells (SOFC)

A solid oxide fuel cell (SOFC) shows highly efficient conversion of carbon-based fuels to electricity, arising as a promising alternative for a wide range of applications, especially in the case of power generation in stationary plants. These devices can provide up to several MW [4]. In particular, SOFC technology has two main characteristics: the electrolyte and electrodes are solid, and versatility in the choice of fuels and oxidants in comparison with most of the existing fuel cells. This versatility also allows the presence of compounds such as CO (considered a poison in fuel cells that operate at lower temperature, e.g. PEMFCs) and the possible use of hydrocarbons (mainly methane) as the fuel. Although the operating temperature is relatively high (650-1000 ºC), which greatly increases costs, they have a relevant number of advantages. Among them, it is highlighted the possibility of taking advantage of the heat for co-generation. In addition, these fuel cells eliminate noise usually associated with power generation systems and produce no or very low levels of SO_x and NO_x emissions [5].

SOFCs are relatively simple devices, since they essentially consist of two porous electrodes (anode and cathode) separated by a dense oxygen ion-conducting ceramic membrane as the electrolyte [5]. The membrane can be a structural component (thickness above 100 µm) or it can be supported on porous anode or cathode structures (5–50 µm) [6]. Using bipolar plates as interconnectors, single cells are connected in series to constitute a stack. Thus, interconnectors are responsible for connecting the cathode of a cell with the anode of the next one. Figure 1 illustrates the scheme described above differentiating between tubular and planar geometries. Tubular designs are more robust while planar cells support the set of cells on a ductile metallic support structure.

In the last years, a common set of materials has emerged to satisfy the requirements of electrochemical performance and stability within the process of SOFC development. Initially, SOFCs were developed for operation primarily in the range of 900-1000 ºC, being this temperature range a problem for suitable materials. However, the reduction of the operating temperature by 200 ºC or more involves a key advance, simplifying thermal management, aiding in faster start-up and cooling and resulting in less degradation of the cell and stack components. These advantages allow the use of a broader set of common materials as summarised in Table 1 [4]. Besides, the material composition may vary depending on the SOFC stack design (Table 2) [7]. The dominant material comes from the layer providing structural support. This can be any functional layer (electrolyte, anode, cathode or interconnect) or an external substrate.
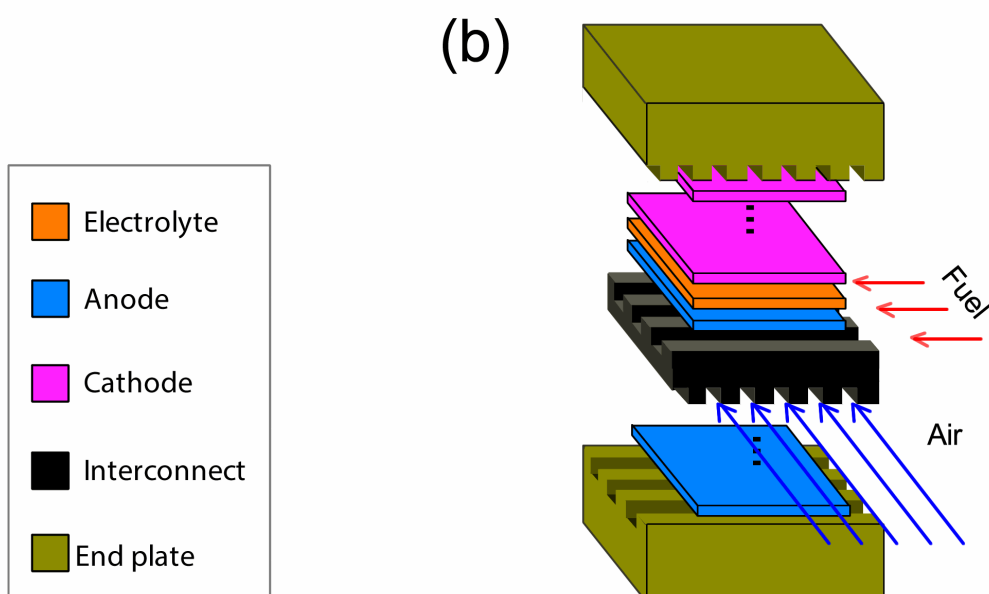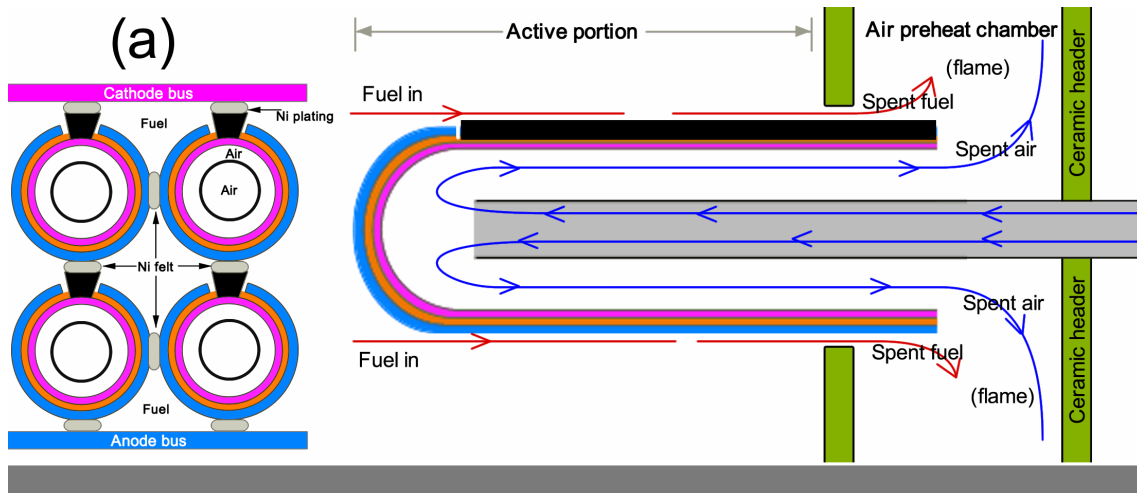
Figure 1. Designs of the SOFC stack: (a) tubular SOFC stack, and (b) planar SOFC stack.

Table 1. Common SOFC stack materials (based on [4]).

Component	Material	Material classification	Material value
Electrolyte	Yttria-stabilised zirconia	Non-hazardous	Medium
Anode	Nickel oxide	Hazardous (cat. 1 carcinogen)	Medium
	Nickel	Hazardous (cat. 1 carcinogen)	Medium
Cathode	Strontium-doped lanthanum chromate	Hazardous (irritant)	Medium
Interconnect	Doped lanthanum chromate	Hazardous (irritant, harmful)	Medium
	Inert metals/alloys	Non-hazardous	High
Sealant	Glass/glass-ceramic	Non-hazardous	Low
Substrate	Ceramic	Non-hazardous	Low

Table 2. Material composition for tubular and planar SOFC concepts (based on [7]).

Material	Tubular concept (wt%)	Planar concept (wt%)
Yttria-stabilized zirconia	3	19
Nickel oxide	3	1
Strontium-doped lanthanum manganite	93	1
Doped lanthanum chromite	1	N/A
Inert metals/alloys	N/A	79

Among the potential materials to be used as electrolytes in SOFCs, yttria-stabilised ZrO_2 (YSZ) is highlighted due to its higher stability at high temperatures during long operating times [5,6]. However, working at high temperatures significantly increases the costs of these devices because of restrictive conditions on the other components. The list of feasible materials that meet all the requirements needed for the operation in the anode of a SOFC is not very wide. In this respect, dozens of research groups worldwide focus their efforts on the search for new materials. The excellent catalytic properties of Pt or other precious metals (such as Au) make them potential candidates. However their high costs, added to the costs of structural materials, have led to the search for other materials. For instance, Ni is a catalyst with adequate performance and the composite Ni-YSZ anode provides better adhesion of the anode to the electrolyte. In a Ni-YSZ anode, nickel plays the dual role of catalyst for hydrogen oxidation and electrical current collector. For these reasons, Ni is found as the most commonly used catalyst in SOFC stacks.

At present, the most used cathode materials are perovskite-family oxides. In this case, the preferred candidate is lanthanum manganite ($LaMnO_3$), which has superior p-type electronic conduction due to the formation of large amounts of Mn^{4+} [6]. Moreover, doped $LaMnO_3$ presents adequate electrocatalytic activity for oxygen reduction. Regarding interconnect materials, lanthanum chromite ($LaCrO_3$) derivatives are the most currently used materials. However, it should be noted that a reduction in working temperature (below 700 °C) would involve the possibility of using stainless steel, thereby significantly reducing manufacturing costs.

Finally, successful development of sealing materials, especially in the planar SOFC design, is critical. The use of appropriate sealing materials largely influences the long-term performance stability and lifetime of planar SOFC stacks and hence their commercialisation at competitive costs. The role of sealing within the stack is key in order to prevent, or at least minimise, leakage of both fuel and oxidant gases from the stack to the environment. Rigid seals typically rely on a wide variety of glass and glass-ceramic materials, including alkaline silicate, alkaline earth silicate, borosilicate, aluminoborosilicate, and others. These materials stick the adjacent stack components together during stack manufacturing (at a temperature above the operating temperature), and then become rigid and immobile when cooled to the operating temperature. Corrosion of stack components and matching thermal expansion (thermal cyclability) are still challenges for sealing.

4. Description and components of Proton Exchange Membrane Fuel Cells (PEMFC) and Water Electrolysers (PEMWE)

A fuel cell is an electrochemical device that converts directly the energy content of a fuel into electricity through a reduction–oxidation reaction. Fuel cell stacks include an anode compartment receiving the fuel whose oxidation yields electrons. The electrons reach a cathode compartment through an external circuit, and they participate in the oxidant reduction. Anode and cathode compartments are separated by an electrolyte, which is in charge of ions distribution.

Electrolysers operate in reverse, i.e. they involve the same components but using electricity to split molecules. In water electrolysis, electricity is supplied to split water molecules into hydrogen (at the cathode) and oxygen (at the anode).

Typically, the parameter used to classify fuel cells and electrolysers is the electrolyte on which their stack is based. In a PEM stack, the main components are the polymeric membrane and the anode and cathode compartments (Figure 2). Although the list of components that characterises PEMFCs and PEMWEs devices is similar, their composition may vary due to different operating conditions. In the case of PEMWEs, the high overvoltage imposed at the anode side and the high concentration of oxygen would promote the oxidation of carbon or graphite materials to CO_2. Therefore, these devices primarily use metallic components (porous structures, flow fields and separator plates) made from Ti, Ti-alloys or coated stainless steel. Tables 3 and 4 summarise common materials used in the manufacturing of a PEMFC and PEMWE stack, respectively.
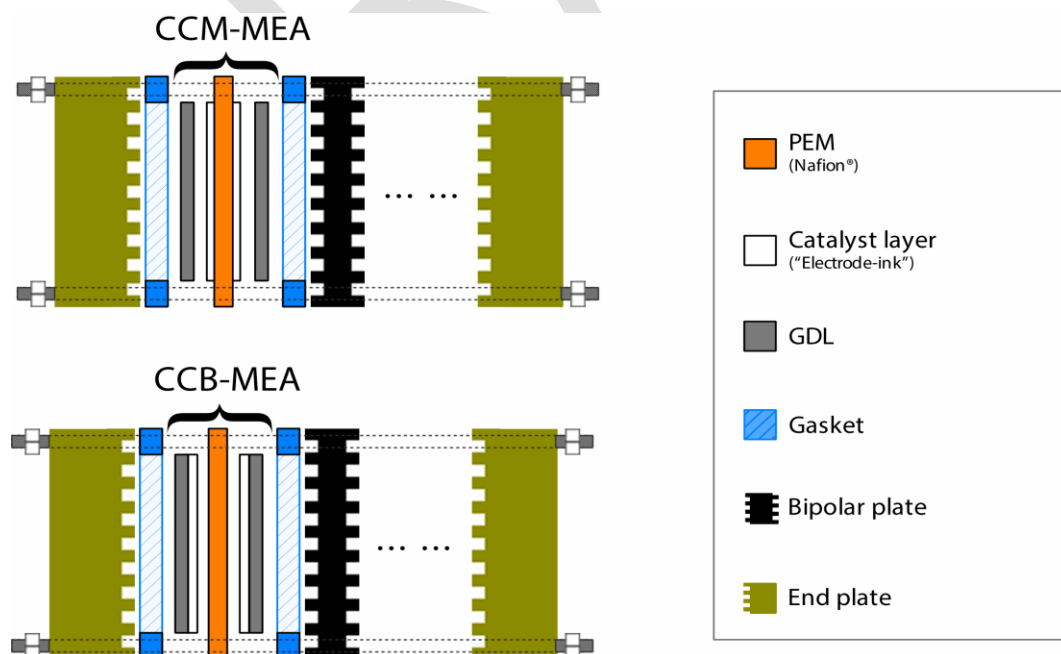

Figure 2. PEM stack concepts and main components.

Table 3. Common PEMFC stack materials.

Component	Material	Material classification	Material value
Electrolyte	Perfluoro-sulphonic acid (Nafion® type)	Non-hazardous	Medium
Anode and cathode - GDL	Carbon-based material (cloth, felt, foam or paper) treated with hydrophobic acid	Non-hazardous	Low
	Metallic mesh or cloth (e.g. stainless steel)	Non-hazardous	Low
Anode and cathode – catalyst layer	Platinum/Pt-alloys	Non-hazardous	High
	Catalyst support (carbon, metal oxides, carbides, etc.)	Non-hazardous	Low
Interconnect	Graphite/carbon-polymer composites	Non-hazardous	Low
	Aluminum/metal alloys	Non-hazardous	High
Sealant	Plastics	Non-hazardous	Low
	Elastomer	Non-hazardous	Low

Table 4. Common PEMWE stack materials.

Component	Material	Material classification	Material value
Electrolyte	Perfluoro-sulphonic acid (Nafion® type)	Non-hazardous	Medium
Anode and cathode - GDL	Thermally sintered Ti	Non-hazardous	Low
	Ti/stainless steel	Non-hazardous	Low
	Graphite/graphite composites (only possible on cathode side)	Non-hazardous	Low
Cathode – catalyst layer	Platinum/Pt-alloys	Non-hazardous	High
Anode – catalyst layer	Iridium/Ir-alloys	Hazardous (irritant, harmful)	High
	Ruthenium/Ru-alloys	Hazardous (irritant, carcinogen)	Medium
Interconnect	Coated Ti/Ti-alloys	Non-hazardous	Low
Sealant	Plastics	Non-hazardous	Low
	Elastomer	Non-hazardous	Low

The stack involves the membrane electrode assembly (MEA) along with collectors and bipolar plates. In the structure of a single cell, the cathodic compartment is separated from the anodic one by the polymeric electrolyte. The electrolyte is commonly a perfluoro-sulphonic acid membrane (Nafion® type) with 20-250 µm thickness, electrically insulated but proton permeable. Because the membrane has to be kept humid to improve its proton conductivity, heat and water management systems are critical parts to ensure optimal operating conditions.

Regarding MEA, there are two typical assembly approaches, which are shown in Figure 2. On the one hand, in the case of the three-layer MEA or catalyst-coated membrane (CCM), the electrolyte membrane is coated on both sides with a catalyst layer. A gas diffusion layer (GDL) with 0.1-0.5 mm thickness is then assembled to each side of the CCM. On the other hand, in the catalyst-coated backing

(CCB) concept, the catalyst layers are supported on the GDLs. In both options, the MEA is the core of the stack, in which the electrochemical reactions take place (i.e., hydrogen oxidation and oxygen reduction).

Extensive research has been made to minimise the cost of PEM devices through a sound selection of materials. In this regard, platinum is the main electrocatalyst used in PEM stacks. Significant efforts have been made to decrease Pt loading from more than 1 g Pt/kW in the past decade to ca. 0.5 g Pt/kW nowadays, mainly in the transport sector in order to make PEMFC vehicles competitive in comparison with battery or internal combustion vehicles [8]. The target for the next decade, set by the U.S. Department of Energy, is 0.1 g Pt/kW [9]. Novel approaches focus on the use of Pt-free compounds (non-noble metals, alloyed transition metals, alternative noble metals), for which low-price and long-term stability are the main requirements. It should be noted that, in the case of PEMWEs, ruthenium and iridium show the best catalytic properties for the oxygen evolution reaction, but iridium is preferred due to greater corrosion resistance.

The GDL is a layer of porous and electrically-conductive material (pressed carbon fibres) externally adjacent to the two electrodes. GDL performs the functions of reactant transport and heat/water management, mechanical support to the MEA, and protection of the catalyst layer from corrosion or erosion. The GDL may be composed of a single layer of carbon-based material (cloth, felt, foam or paper), and it is often treated with polytetrafluoroethylene (PTFE, 10-40 wt%) to avoid water flooding. A novel approach is the assembly of the MEA structure using dual-layer GDLs, preferably following the CCB-MEA concept. The external layer, characterised by macro-porosity, performs the function of gas diffusion and mechanical support while the internal layer, treated with PTFE and contiguous to the catalyst layer, reduces the contact resistance and enhances water management in the MEA [10].

To achieve the desired electric power of a stack, multiple MEAs are assembled in series interconnected by bipolar plates. A bipolar plate holds the function of electrical collector between two adjacent cells of a stack. The materials generally used for bipolar plates are graphite, polymeric compounds or carbon-polymer composites. Aluminium and metal alloys such as Cr-Ni steel are also considered suitable given their high electrical conductivity and good mechanical properties. However, due to corrosion concerns, metallic bipolar plates need to be treated or coated on their surface [11]. The surface of bipolar plates presents a set of channels that have the double function of reactant delivery and coolant handling. In order to prevent potential leaks and ensure an optimal compression between the sandwiched layers, gaskets (made up of silicon, Teflon®, rubber, etc.) are interposed.

The PEM stack is completed by two end plates. They provide uniform pressure distribution between the layers and ensure a correct sealing. The performance and the lifespan of a PEM stack is highly affected by these aspects [12]. Besides electrical insulation and rigidity, the requirements of end

plates include thermal stability and corrosion resistance due to the operating temperature and the presence of humidity. The typical material used is aluminium.

PEMFCs are seen suitable for a large number of applications, ranging from micro fuel cells to large stationary applications and including use in transportation in light-duty and bus fleets. Due to the high intolerance of the catalyst (Pt) to CO (< 20 ppm), very high-purity hydrogen is needed when using PEMFCs. Generally, the operating temperature is in the range 70-80ºC (low-temperature PEMFCs) while PEMFCs operating at higher temperature (up to 180ºC) are under development. The increase in operating temperature allows a higher CO tolerance, the reduction of catalyst load and more efficient heat removal, but information about the performance of high-temperature PEMFCs is still scarce.

5. Description and components of Alkaline Water Electrolysers (AWE)

The AWE technology is a mature technology which includes most of the installed water electrolysis capacity worldwide. The power range that commercial systems are able to provide varies from 1.8 to 5,300 kW with associated investment costs in the range of 900-4,500 €/kW depending on the production capacity [13]. This wide range of power makes current AWE systems the most suitable option for large-scale applications, with hydrogen production rates for commercial systems between 0.25 and 760 Nm³/h [14]. The purity of the hydrogen obtained can be higher than 99.9% without auxiliary purification equipment. In the last years, efforts have been focused on improving the efficiency of AWEs (currently between 73 and 83%) in order to reduce the operating costs associated with electricity consumption (4.5-7 kWh/Nm³ H_2) [13,15]. The lifetime of the AWE system is approximately 30 years although inspections are required every 7 to 15 years with the aim of replacing or reactivating the electrodes and the diaphragms.

The operating principle of an AWE cell is shown in Figure 3. The electric field established by an external power source decomposes water molecules and the diaphragm allows passing hydroxide ions from the cathode to the anode. Hydrogen is formed at the cathode, and oxygen at the anode through the recombination of hydroxide anions. The following reactions take place in an AWE cell:

Anode: $4OH^- \rightarrow O_2 + 2H_2O + 4e^-$

Cathode: $4H_2O \rightarrow 2H_2 + 4OH^- - 4e^-$

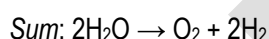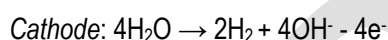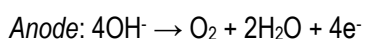*Sum*: $2H_2O \rightarrow O_2 + 2H_2$

Table 3 summarises common materials used in the manufacturing of an AWE stack. The AWE cell contains two electrodes separated by a gas-tight diaphragm which is submerged in the electrolyte. Generally, the electrolyte is one of the most characteristic components of the water electrolysis cell. The electrolyte is usually a 20-40 wt% aqueous solution of potassium hydroxide, which is preferred over sodium hydroxide (KOH) due to higher ionic conductivity [13]. However, the KOH electrolyte also presents a highly corrosive nature, which leads to use only a limited number of materials in the AWE system. Metal sheets, e.g. nickel or stainless steel, or plastics such as acrylic materials, PTFE or Plexiglas® can be used.

One of the main advantages of AWE systems in comparison with other types of water electrolysers is the relatively low cost of the electrode materials. In this sense, a high number of publications on AWE technology describe the use of relatively low-cost electrode materials such as nickel or copper for both the anode and the cathode [15]. The type of material directly affects the exact kinetics of the reactions that take place in the electrodes. As well, the performance of the catalysts depends not only on the catalyst material, but also on other factors such as the support material. A number of catalysts

for AWE systems are currently available or under development, but Ni-based electrodes arise as the best candidates due to the reduced corrosiveness of the produced oxygen and their reduced costs.

Cathode catalysts can be classified into three main groups: metals with improved microstructure and surface area, metal alloys, and hydrogen storage alloys [15]. The first two groups include Ni- and Raney Ni-based catalysts, respectively, which are widely recognised as the best cathode candidates in the AWE technology. Ni-based catalysts can achieve high stability and activity by coating with or co-depositing a more active or more stable material such as cobalt, aluminium or iron. Catalysts based on Raney-Ni (solid catalyst composed of fine grains of a Ni-Al alloy) doped with elements such as Fe, Cr, Mo, Sn, or Co have also been shown as appropriate candidates, with several advantages compared to pure Ni electrodes. According to relevant literature, the best long-term performance can be achieved with 70% Raney-Ni, 10% carbonyl Fe, 10% carbonyl Ni and 10% PTFE (binder) [15]. Hydrogen storage alloys present a lower interest for their use as cathode catalyst in AWE systems. With respect to anode catalysts, two main common groups are found: metal-based catalysts, and oxide-based catalysts. In particular, precious metals such as Pt, Rh, Ag or Pd-based catalysts and their alloys with nickel show suitable performances.

The diaphragm is also a key component to take into account within the EoL strategy of AWE systems. The main requirements for the membrane are: gas separation, electrical isolation, and high chemical and thermal stability. Following these criteria, the diaphragm is typically porous using inorganic, ceramic or, recently, organic materials. For instance, PTFE, low-density polyethylene and polybenzimidazole doped with an anion-exchange phase arise as feasible candidates for the diaphragm. Besides, common plastics such as polyethylene or epoxy resin are used to provide a frame around the electrode (with holes drilled for gas delivery), and polycarbonate/acrylonitrile butadiene styrene or PTFE are generally used as the separator between cells, replacing the graphite or metal bipolar plate used in bipolar stacks. Finally, a current collector in the form of a copper sheet and welded onto one edge of a wire mesh (support for electrodes made up of nickel or copper) is required.

Within the AWE system, it is necessary to maintain two liquid streams: liquid electrolyte, and water. Assuming no physical losses, the liquid electrolyte is not consumed, being pumped back to the cell in order to maintain its concentration. However, water must be continually supplied in order to maintain the desired electrolyte concentration in the cell. Product gases that leave the cell (i.e., hydrogen and oxygen) are separated from the remaining electrolyte and separately stored in gas separator tanks. The full electrolytic process is illustrated in Figure 3 (including not only components of the AWE stack, but also components corresponding to the BoP).
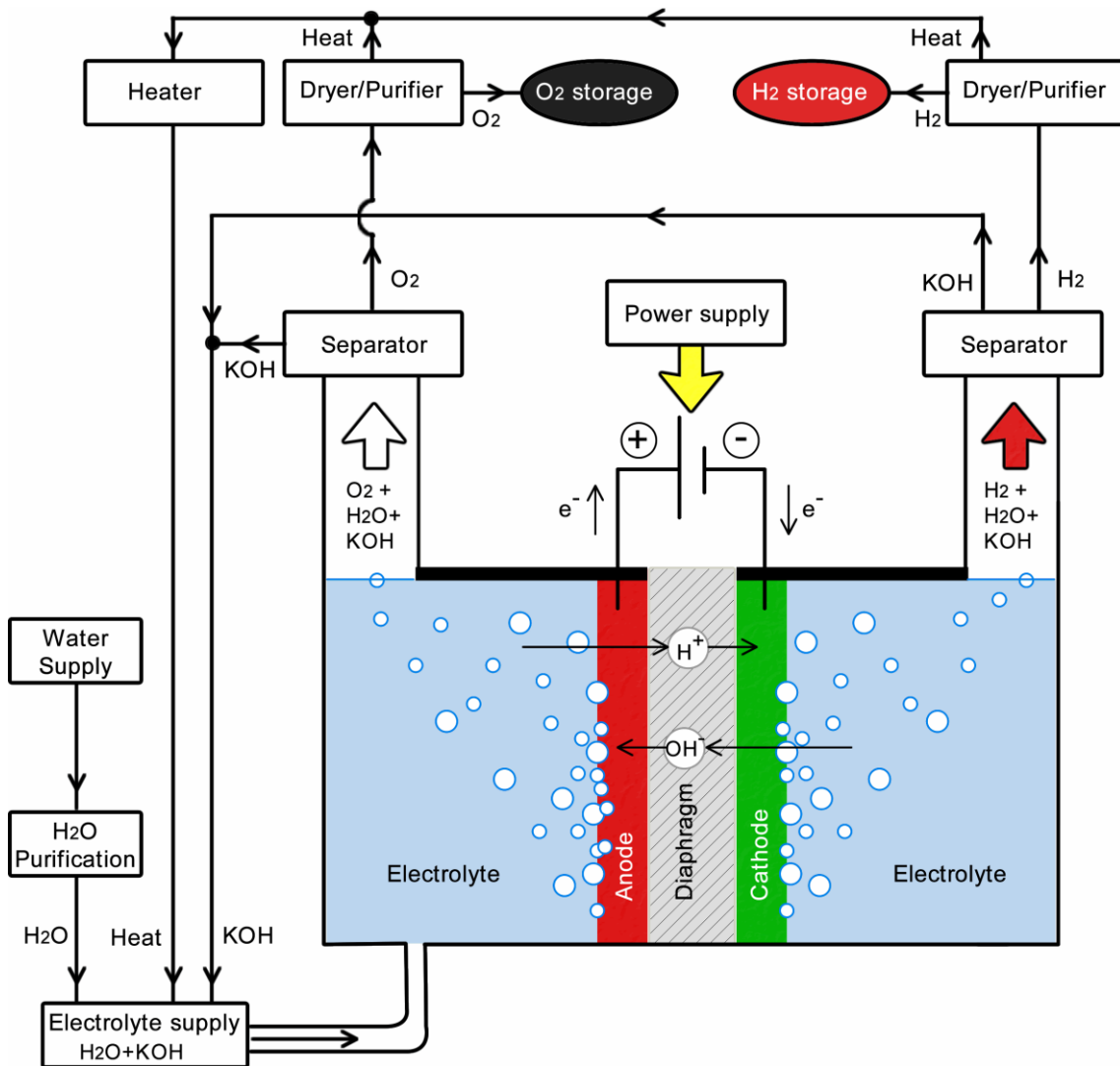
Figure 3. Operation of an AWE cell.

Table 5. Common AWE stack materials (based on [15]).

Component	Material	Material classification	Material value
Electrolyte	Potassium hydroxide	Hazardous (corrosive)	Medium
Anode	Precious metals	Non-hazardous	High
	Plastic	Non-hazardous	Low
Cathode	Raney-Nickel	Hazardous (cat. 1 carcinogen)	Medium
	Plastic	Non-hazardous	Low
Diaphragm	PTFE/Polymers	Non-hazardous	Low
Interconnect	Plastic	Non-hazardous	Low
Sealant	Plastic	Non-hazardous	Low

6. Description and components of Balance-of-Plant (BoP) equipment

Regarding the BoP, general elements common to all types of stacks (power conditioning system, current collectors, air/fuel supply infrastructure, heat and water management system, electronic components, start-up and shut-down systems) are distinguished from components that are specific to the application of the device or dependent on hydrogen purity. Table 4 summarises the main components of the BoP. The size of the BoP is proportional to the power of the stack, which varies from less than 1 W to more than 1 MW. A critical aspect of BoP components is their lifespan, especially for those components with moving mechanical parts (e.g., supply compressors or circulating pumps) whose materials have to guarantee a friction as low as possible in order to minimise the degradation rate.

Table 6. Main components and materials of BoP.

BoP components	Main materials
Blower or compressor	Metals, plastics
Humidification membrane	Metals, plastics, polymers
Pumps	Metals, Teflon®, rubbers, plastics
Regulators	Metals, plastics, rubbers
Printed circuit boards (PCBs)	Metals, plastics, semiconductors, precious metals
Power conditioning system	Metals, plastics, semiconductors, precious metals
Deionising filter	Metals, plastics, resins
Pipes	Metals, plastics, rubbers
Valves	Metals, plastics, nylon, Teflon®
Gaskets (piping system)	Paper, plastics, rubbers
Thermal insulation system	Mineral wool, fibreglass
Heat exchangers	Metals
Sensors	Plastics, precious metals, semiconductors, glass
Water condensers	Stainless steel

The thermal insulation system can be considered a part of the BoP. It is a crucial part of the whole system and its materials need special attention, particularly in high-temperature fuel cells (SOFC), in which thermal stress and fatigue are severe. Besides thermal insulation and resistance, other requirements for these materials are low weight, easy shaping and non-combustibility. Regarding the insulation system for PEM stacks, typical solutions are porous foam layers and polymeric panels filled with inert gases. Moreover, being fuel cells direct-current devices producing low-voltage and high-current power, high thermal losses may occur through the external current collectors. As thermal resistance losses are proportional to electrical resistance, high-conductivity materials such as copper or silver can be used in the current collectors to reduce these losses, or collectors with larger cross section using materials with lower conductivity such as stainless steel.

In PEM devices, as the membrane has to be kept humid, a critical component of the BoP is the humidification system. The need for humidification in low-temperature PEM stacks increases the general complexity of the system. One of the requirements of the humidification system relates to the purity of water, which has to be deionised. To optimise input consumption, water for humidification is internally

recovered, thus avoiding an external circuit. An efficient water management system increases significantly the performance of the stack.

Advanced polymers are suitable materials for BoP components of low-temperature fuel cells, but stainless steel alloys are preferred materials for the BoP of SOFCs due to the high operating temperatures. In SOFC systems, the main differences with the rest of fuel cell technologies lie on the fuel delivery subsystem and the materials used for thermal insulation [4,6]. Regarding the fuel supply subsystem, integrated and direct internal reforming are the only options for high-temperature fuel cells. In particular, direct internal steam reforming is preferred in SOFC systems. Small amount of noble metals such as Pd, Ru, Pt or Ni are generally used as promoters to enhance the catalyst's performance in SOFC reformer subsystems. Both cases require pre-reforming of the hydrocarbons (at 300-450 ºC) to a methane-rich fuel in order to avoid deposition in the fuel feeding pipes and heat exchanger. High-alloy steels and carbon composites as well as alumina alloys are usually chosen as suitable materials for the fuel delivery subsystem [6].

Materials used in high-temperature fuel cells for thermal insulation require: high thermal transfer resistance, high temperature resistance (up to 1000 ºC), thermal shock resistance, easiness to shape, no health hazard, non-combustibility, light weight, and low cost. Microporous and aerogel insulation are found to be appropriate candidates for meeting these requirements with currently decreasing costs. Most aerogels are silica based, but high alumina content materials are also available. The insulation is available in panels and spray-on versions [6]. Another less expensive option is the use of mineral wool for thermal insulation.

7. EoL technologies

In general, the success of products is highly conditioned by their design, which must take into account EoL strategies according to the principles of eco-design or design for recycling [7,16,17]. Nevertheless, all the strategies of design for recycling are not always possible, as they may occasionally bring about negative effects on the technical performance or the final cost of the product [17,18]. Recycling compatibility charts can be used for the selection of materials in order to allow practical recovery processes for critical materials (or even components). Recycling compatibility charts are commonly used during design to determine the possible joint recyclability of materials. Other examples include the use of materials with different density or magnetic properties in order to facilitate their separation, and the reduced use of painting to avoid the contamination of materials, especially plastics. For instance, metals usually mean more than 75 wt% of fuel cell systems [19]. The interest of recycling companies focuses mainly on steel –for which mature recovery technologies are available– and noble metals used as electrocatalysts. However, the rate of metal recovery would be compromised if the rules of compatibility are not considered during the design, e.g. aluminium recyclability would be affected by contamination with zinc, iron and/or copper. Unlike recovery methods for iron and steel, the technologies for recovering spent catalysts present large room for improvement, which makes them highly attractive for both research and industry actors. In this respect, catalyst recovery would lead to a significant reduction in the costs of FCH technologies, also mitigating scarcity concerns.

The components and materials identified in FCH products can be classified into hazardous materials and high and low economic value ones. In this sense, a hazardous material is a waste with properties that make it dangerous or capable of having a harmful effect on human health or the environment. The recovery of high-value and hazardous materials is a priority in EoL strategies. High-value materials should be reused or recycled in a valuable application, while the recovered hazardous materials (depending on purity) may be suitable for high- or low-value application. Finally, the disposal (landfilling) of low-value materials should be avoided, prioritising their reuse or recycling.

EoL strategies for FCH products –as waste electrical and electronic equipment (WEEE)– present common steps (Figure 4). After collection of the device, hardware components have to be separated and divided into groups of compatible materials for subsequent joint processing. Manual disassembly, during which all fluids have to be removed, facilitates the potential reuse and remanufacturing of components as commonly preferred options that lead to savings in terms of energy, natural resources, costs and landfilling space. For instance, in AWEs, the spent KOH solution is separated from solid materials and neutralised. In order to enable manual disassembly, the component accessibility and disconnectability should be maximised during the design of the system, also taking into account volume constraints. After manual disassembly, separate strategies for stack and BoP components and

materials can be drawn. Size reduction (grinding, crushing and pulverisation) and mechanical separation steps usually take place when dealing with both BoP and stack components.
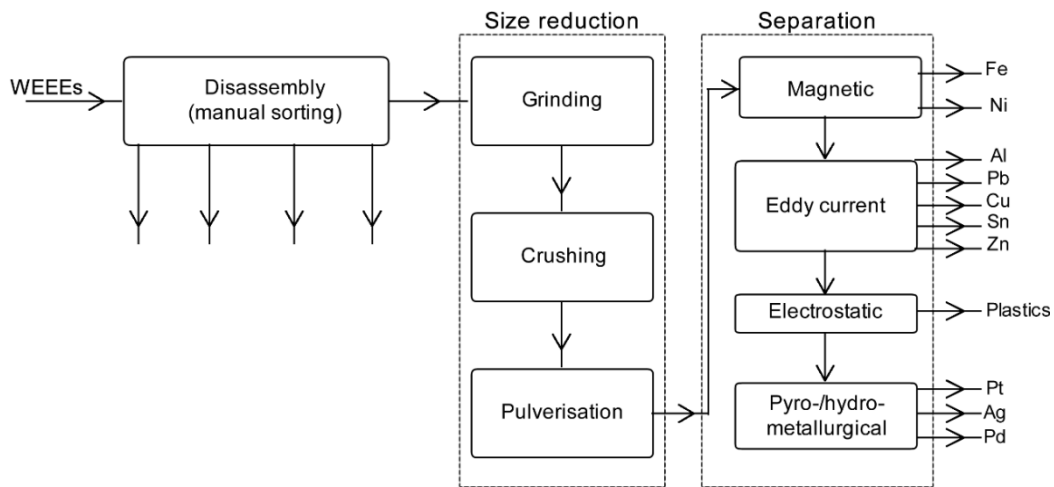

Figure 4. Common steps in EoL strategies.

Regarding the BoP, Table 5 presents potential EoL strategies based on the guidelines provided by the Society of Automotive Engineers [18]. Potential steps concerning purification and storage of the primary fuel are considered to be outside the boundaries of the system.

Table 7. Potential EoL strategies for BoP components (based on [18]).

BoP components	Main Materials	EoL strategy
Blower or compressor	Metals, plastics	Reuse/material recycling
Humidification membrane	Metals, plastics, polymers	Reuse
Pumps	Metals, Teflon®, rubbers, plastics	Reuse
Regulators	Metals, plastics, rubbers	Reuse/alternative use
PCBs	Metals, plastics, semiconductors, precious metals	Material recycling
Power conditioning system	Metals, plastics, semiconductors, precious metals	Material recycling
Deionising filter	Metals, plastics, resins	Reuse
Pipes	Metals, plastics, rubbers	Reuse/material recycling
Valves	Metals, plastics, nylon, Teflon®	Reuse/material recycling
Gaskets (piping system)	Paper, plastics, rubbers	Material recycling/energy recovery
Thermal insulation system	Mineral wool, fibreglass	Energy recovery
Heat exchangers	Metals	Reuse/material recycling
Sensors	Plastics, precious metals, semiconductors, glass	Material recycling
Water condensers	Stainless steel	Reuse/material recycling

7.1 Treatment of common metals

Given the existing infrastructure for metal recycling, high recovery rates can be achieved. In this sense, aluminium, copper and steel parts can be recycled in high rate by following conventional routes (through several steps of mechanical treatment such as size reduction and separation). Although titanium is a precious metal, its recycling process fits the EoL strategy presented in this section. Initial material separation is carried out by means of mechanical separation. Ferrous metals are recovered from the residue using magnetic separation, and eddy current separation is used to recover non-ferrous metals.

The housing components of the devices can be recycled for material recovery before the mechanical processes, or even reused after reconditioning.

Metal recycling is a normal economic practice. For instance, about 75% of all copper-based products are made from recycled copper [15]. Metals can generally be processed many times with no quality loss. First, when metal scrap is received for recycling, it is inspected and graded. The scrap material is melted and, in some cases, brought to higher purity while melted. Regarding metal alloys, they have to be segregated, kept clean and identified so that the content in alloying elements and impurities can be known. Metal alloys are then melted together into batches of known composition, some with virgin material in order that the recycled material presents the desired alloy composition.

7.2 Treatment of common plastics

Within the whole EoL strategy for FCH products, the recovery of polymer materials presents relatively low priority. However, the generation of high amounts of polymer waste (especially plastic waste) worldwide is a complex issue that requires the implementation of suitable, feasible and environmentally-friendly recycling processes.

PTFE is one of the main polymer materials found in the FCH products considered. PTFE can be recycled into other products and used e.g. for tubing and sheets. PTFE recycling became common in industries because no chemical reaction is required. Although different compositions can be found, the PTFE recycling process is similar to that for copper. The PTFE scrap is ground into fine powder and then blended with pure PTFE. To remove the inorganic compounds, the scrap is heated before grinding. Only the extruded forms can be recycled and separated from undesirable impurities. It is then put into a long strand, which is subsequently cut down into small pieces and sent to industries to be used as recycled material for their products.

The rest of polymer waste streams must also be processed, which may involve a wide range of recycling and recovery activities [16]. EoL strategies for waste reprocessing into valuable products are seen as the most suitable strategies for plastic waste management. If no reprocessing treatments could be implemented, energy recovery processes may be suitable but with lower priority within the recommendations of the HyTechCycling project. Once the polymer waste is correctly pre-treated, mechanical recycling or thermal treatment can be applied. In the first recovery strategy, plastic waste is reshaped to obtain new marketable products. In order to achieve this goal, different transformation techniques are possible. Granulation, extrusion and injection moulding are suggested as viable solutions. The second EoL strategy (incineration) can be used to recover some of the energy content of the plastic. The useful energy recovered can vary considerably depending on whether it is used for electricity generation. Even though plastic waste incineration reduces landfilling needs, it is important to take into

account the environmental concerns associated with the emission of hazardous substances into the atmosphere during the process.

7.3 Treatment of precious metals

Pyrometallurgical and/or hydrometallurgical processes are conventionally used to recover precious metals in a refining stage [20,21]. The initial content of precious metals and their recovery efficiency [22,23] determine the profitability of the process, which varies with the refining process and the type of equipment.

For instance, the concentration of precious metals in PCBs –common components in many types of WEEE– is much higher than in natural ores [24]. Being rich in copper and precious metals such as gold, silver and palladium, the interest of research and industry actors in PCBs and their EoL is increasing. A number of methods based on hydrometallurgical and/or pyrometallurgical processing are studied for cost-competitive recovery of precious metals from PCBs [24–27]. Disassembly is usually the first stage, in which hazardous components such as batteries and condensers are separated and treated independently. The next step means crushing PCBs into micro-pieces through grinding or shredding. Afterwards, PCBs in the form of powder undergo a series of separation steps according to magnetic, size or density properties in order to recover valuable metals and non-metals.

7.3.1 Hydrometallurgical processes

The hydrometallurgical pathway involves the dissolution of interesting elements from solid matrices through acid or caustic attacks followed by separation via solvent extraction, precipitation, cementation, ion exchange, filtration or distillation. Strengths of this family of methods include reduced risk of air pollution, higher selectivity of materials (metals), lower energy consumption and the possibility to reuse chemical reagents. On the other hand, drawbacks include the need for mechanical pre-treatment to reduce the volume and increase the effective surface exposed to the chemical agents, the large volume of solutions, and the generation of wastewater, which may be corrosive and/or toxic.

7.3.1.1 Platinum-group metals recovery

EoL processes for FCH products often focus on the recovery of platinum-group metals (PGM) such as platinum, iridium or ruthenium, given their economic relevance. For instance, above 45% of the total cost of PEM systems can be attributed to the electrodes [28]. Hydrometallurgical processes to recover PGM from the catalyst layer of PEM stacks (fuel cells or electrolysers) or from the spent catalyst of SOFC reformers involve leaching, separation and purification sub-processes. Further sub-processes (e.g., regeneration of solvents) may be involved in order to improve the economic and environmental performance of the whole process [29,30]. In the case of PEM systems, a pre-treatment to delaminate the different layers of the MEA is necessary. Although the following steps presented in this section focus on platinum recovery, they are valid for the recovery of PGM (e.g., iridium and ruthenium).

Leaching of spent catalysts

The leaching agent is a mixture of a strong acid and an oxidant. *Aqua regia* is typically used for the leaching of spent catalysts in the recovery of PGM (on carbon support*)*. It is a solution of hydrochloric acid and nitric acid with molar ratio 3:1, and its use as a leaching agent allows Pt-recovery percentages above 95% [30,31].

The leaching process generates very harsh working conditions due to high acid concentration with pH < 1. Although different acid/oxidant solutions are possible, *aqua regia* is still used in most cases due to high recovery efficiency. After leaching, a filtration stage removes the carbon particles from the PGM-containing solution.

Separation

PGM anions in solution can be separated by different techniques. In particular, for Pt, Duclos et al. [29] investigated two different routes: (i) liquid-liquid extraction followed by platinum stripping, and (ii) anion exchange resin process followed by resin desorption. They considered two different leaching solutions: (i) hydrochloric acid with hydrogen peroxide, and (ii) hydrochloric acid with nitric acid. Conventional liquid-liquid extractants are phosphorus- or ammonium chloride-based. In the study by Duclos et al. [29], they tested two different extractants (a phosphorus-based one [Cyanex® 923] and another based on quaternary ammonium [Aliquat® 336]) and two alternative resins (a quaternary ammonium-based one [Amberjet® 4200] and another based on tertiary ammonium [Lewatit® MP-62]).

Precipitation

In the case of Pt-rich stream coming from the separation process is treated with e.g. ammonium chloride (NH_4Cl) in order to precipitate platinum as $(NH_4)_2PtCl_6$, which is eventually filtered and recovered in solid form as the final product of the process. After filtration, an additional step of ignition at 350 ºC can be conducted in order to obtain high-purity Pt (99.9%) [30]. Figure 5 shows the process described in Duclos et al. [29].
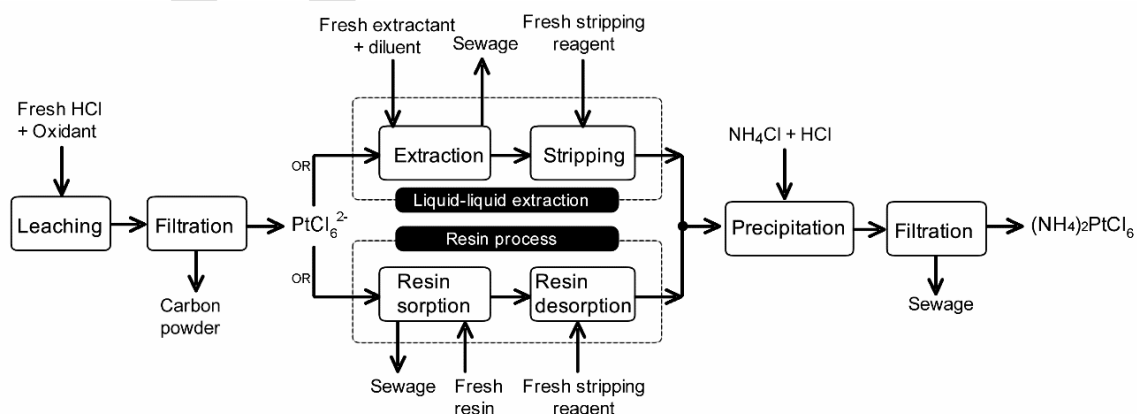

Figure 5. Hydrometallurgical Pt-recovery process (based on [29]).

$(NH_4)_2PtCl_6$ may be reused with carbon powder to generate new electrode ink through different methods (metal sputtering, impregnation reduction, polyols or precipitation reduction). The efficiency of each of the three main stages determines the overall platinum recovery. Experimental results for a PEM system show that, in the best combination, the percentage of Pt recovered from a CCM-MEA configuration is 76% of the initial content [29]. It should be noted that other valuable materials (e.g., Nafion® membrane) are not recoverable when using the hydrometallurgical process described above.

7.3.1.2   Nickel recovery

When following a hydrometallurgical approach, Ni leaching from various sources can be performed using mineral acids or bases. Among the acids used, sulphuric acid shows economic and efficiency advantages.

In AWEs, a hydrometallurgical process is used to treat the spent Raney-Ni catalyst. The process (Figure 6) involves leaching with dilute sulphuric acid to produce nickel sulphate and a low percentage of alumina, followed by purification to produce pure $NiCO_3$ [32]. The spent catalyst is washed with deionised water in a single stage to remove soluble impurities. After filtration, the residue is dried in an oven at 80-90 ºC, obtaining the washed catalyst with 43% Ni and 4% Al. The leaching process is carried out using a H_2SO_4 solution (6-12 vol%). According to the literature, 99% Ni and 39% Al recovery can be achieved using 12 vol% sulphuric acid at 20% solid/liquid ratio, a reaction temperature of 30ºC and 120 min leaching time.
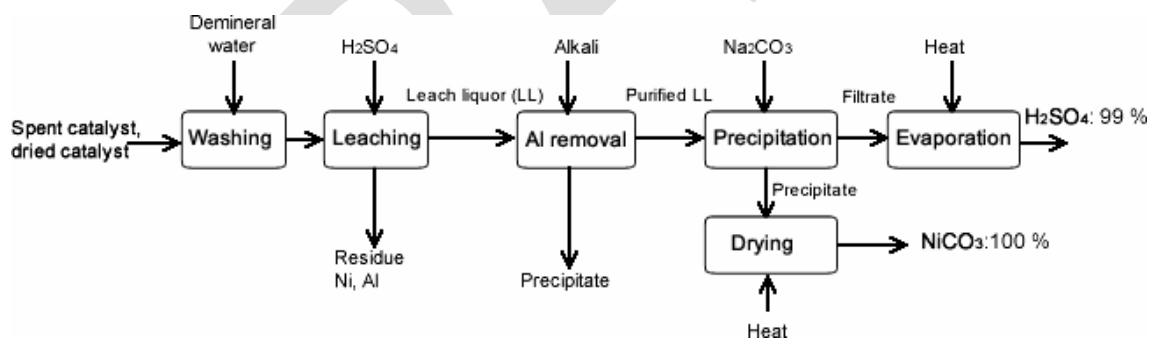

Figure 6. Recovery of nickel carbonate from spent Raney-Ni catalyst (based on [32]).

In SOFCs, the recovery of the anode catalyst is also based on the reaction of nickel oxide with sulphuric acid to form nickel sulphate. A maximum recovery rate of 99% is reached under optimised conditions [33]. Nickel sulphate is a useful feedstock for the electroplating industry, which provides a high-value opportunity for recycling.

7.3.1.3   Silver recovery

This section describes the treatment of waste silver oxide button cells for the recovery of silver metal by smelting and electrolytic routes. Due to similar composition features, this process is relevant to the recovery of silver from the silver oxide spent catalyst in AWEs. The steps required in this process [34]

are shown in Figure 7. The scrap material collected is leached with a nitric acid solution and the temperature is maintained at 30-75 ºC. The solid/liquid ratio is kept at 1:3.4 during leaching. The leached solution is then boiled with a little activated charcoal for removing any organic material. The silver present in the leaching solution is separated as insoluble silver chloride precipitate by adding a 0-15 wt% stoichiometric excess of 25% sodium chloride solution in a slightly acid medium. Then, silver chloride is smelted with sodium carbonate flux to obtain silver metal. Besides, a silver thiosulfate complex electrolyte can be prepared dissolving silver chloride in a sodium thiosulfate solution. This complex catholyte is electrolysed in a polyvinylchloride cylindrical cell divided by a cation exchange membrane. Finally, silver is obtained in the catholyte through an electrolytic step.
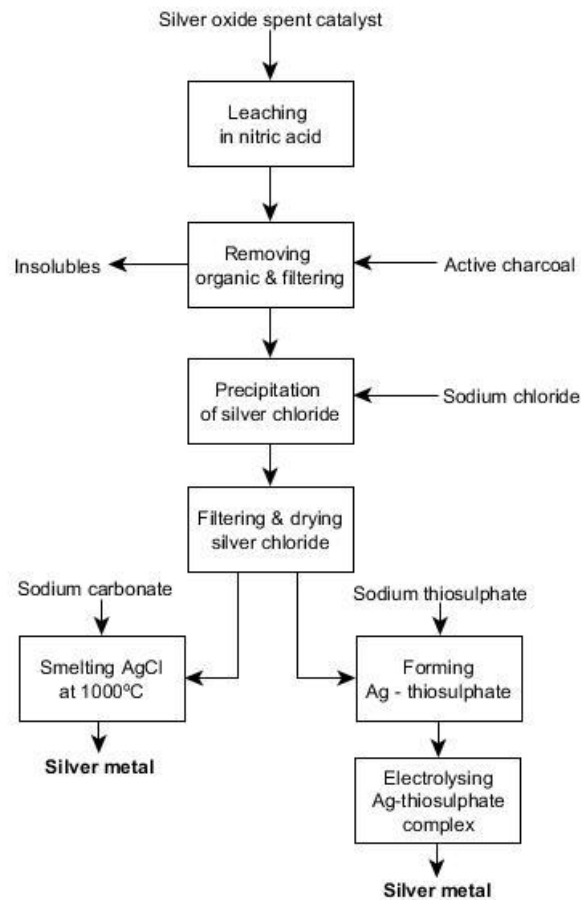

Figure 7. Silver recovery from waste silver oxide button cells (based on [34]).

7.3.2   Pyro-hydrometallurgical processes

In pyro-hydrometallurgy processing, mechanical pre-treatments are not needed. For PEM systems, it involves directly a combustion process in which the membrane, GDLs and electrodes are incinerated. The resulting ashes are processed through dissolution and Pt is then precipitated. When compared to hydrometallurgical processes, pyro-hydrometallurgical process present higher recovery efficiency but higher energy demand.

7.3.2.1 Pt recovery from Pt/C catalyst

Pyrometallurgical methods can be applied to the EoL of PEM stacks to recover only Pt-group metals. In the base case for this type of processes, MEAs are thermally destroyed and Pt is recovered from the residues after their dissolution. A process based on this family of methods is described by Zhao et al. [35], who included the following key stages: drying and calcination, leaching and purification, and reduction and filtration. After the recovering process, Pt is recycled to prepare again the Pt/C catalyst. On the one hand, the simplicity of this type of processes in terms of number of stages facilitates their technical performance. On the other hand, the severe conditions in terms of operating temperature in the first stage (> 600 ºC) and the harsh working conditions in terms of pH represent challenges to overcome concerning safety and environmental issues.

Drying and calcination

In the first stage, the exhausts catalyst of carbon-coated platinum is dried at 80 ºC for 3 hours. Afterwards, calcination is carried out at 600 ºC for 6 hours in order to eliminate the remaining volatile compounds and carbon.
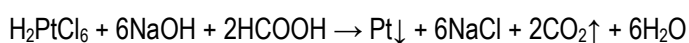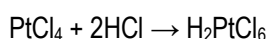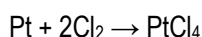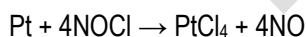
Leaching and purification

The residues of the previous calcination step are fully leached using nitric acid (HNO_3), also known as *aqua fortis*, at 65 ºC. Subsequently, in the purification step, HCl is added to the solution and heated to 110 ºC. The full vaporisation of the solution at room temperature leads to obtain solid H_2PtCl_6 as the Pt/C precursor.

Reduction and filtration

Solid H_2PtCl_6 can be dissolved in deionised water at pH in the range 3-4 adding NaOH. Pt can then be reduced through the addition of methanoic acid (HCOOH).

The main reactions of the process described by Zhao et al. [35] are:

$$3HCl + HNO_3 \rightarrow Cl_2 + NOCl + 2H_2O$$

$$Pt + 4NOCl \rightarrow PtCl_4 + 4NO$$

$$Pt + 2Cl_2 \rightarrow PtCl_4$$

$$PtCl_4 + 2HCl \rightarrow H_2PtCl_6$$

$$H_2PtCl_6 + 6NaOH + 2HCOOH \rightarrow Pt\downarrow + 6NaCl + 2CO_2\uparrow + 6H_2O$$

In the last stage, pure platinum (Pt black) can be recovered through vacuum filtration and drying. Figure 8 shows the main stages of the pyro-hydrometallurgical process described by Zhao et al. [35].

Alternatively, $(NH_4)_2PtCl_6$ can be obtained by treating H_2PtCl_6 with an ammonium salt (e.g., NH_4Cl). Metallic Pt can then be obtained by heating $(NH_4)_2PtCl_6$ in a hydrogen-reducing atmosphere.
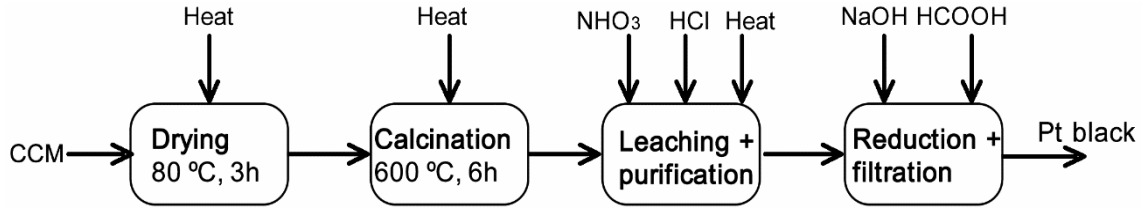

Figure 8. Pyro-hydrometallurgical Pt-recovery process (based on [35]).

7.4 Specific treatments for SOFC systems

Regarding the tubular SOFC concept, the greatest benefits from recycling would be obtained by the recovery and recycling of the cathode material, strontium-doped lanthanum manganite (93% of weight contribution within the tubular SOFC stack), since it is the most contributing material in terms of environmental impact and economic value [7]. This arises as a challenge because it is an uncommon material and no existing recycling process is available. Assuming that the material cannot be substituted for technical reasons, future guidelines and recommendations about the recycling of this key SOFC material within the framework of the HyTechCycling project would be an important contribution to define feasible EoL strategies that support the commercial deployment of SOFC technology.

Concerning the planar SOFC concept, the greatest benefits at the EoL stage would be obtained by recycling the steel interconnect plates, which represent 79% of the total weight of the stack. Besides, due to the weight contribution of YSZ (19 wt%) and its economic value, suitable recovery and recycling strategies should be explored for this material. In this case, hydrothermal treatment is the most extended option for the management of the electrolyte waste. Figure 9 summarises a process proposed in the literature for the recycling of yttria-stabilized tetragonal zirconia polycrystals [36]. The YSZ-containing materials are hydrothermally treated with water at 200-240 ºC under autogenous pressure, leading to their disintegration. The resulting powder is collected by filtration and dried at 50°C. Then it is milled and sieved below 100 µm to remove hard aggregated particles. The sieved powder is uniaxially pressed, which is followed by cold isostatic pressing at 200 MPa. The resulting green body is re-sintered at 1400-1600 °C for 2 h to obtain a recycled YSZ sintered body.
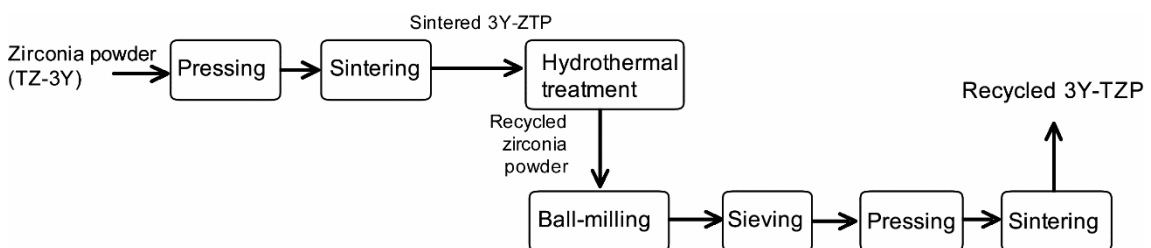

Figure 9. Recovery of YSZ under hydrothermal conditions (based on [36]).

Finally, ceramic materials are the last waste stream found in the EoL of the SOFC stack. The high energy requirements associated with ceramic processing and the inherent low material value do not encourage recycling of this waste stream. The high-temperature environments required during cell fabrication promote the migration of chemical species, and the presence of contaminants –even in trace amounts– leads to performance degradation. Thus, the implementation of ceramic waste recycling processes associated with the SOFC technology is not expected in the near future. Besides, even though high amounts of ceramic waste are currently disposed in landfills, the environmental impacts associated with this waste management suggest avoiding it. Therefore, reuse strategies for SOFC ceramic waste arises as the most suitable practice according to economic and environmental criteria. The construction industry is a potential user of recovered ceramic waste and the use of fired pottery ware in brick manufacture has been reported. Moreover, ceramic material is also a potential replacement for clays and minerals in the manufacture of cement and concrete. Medina et al. [37] showed that ceramic sanitary ware wastes could replace up to 25% of these clays and minerals to obtain concrete for structural purposes. Similarly, Puertas et al. [38] successfully used up to 35% of certain types of ceramic wastes as raw materials for Portland cement clinker production.

7.5 Overall strategy at the FCH-technology level

Figures 10 and 11 show strategic EoL management solutions applied to SOFC systems for tubular and planar designs. Respectively, Figures 12 and 13 show existing EoL strategies applicable to PEM devices making a distinction between two pathways for PGM recovery from the MEA: hydrothermal route and pyro-hydrothermal route, respectively. Finally, Figure 14 presents the EoL strategy followed for AWE devices.
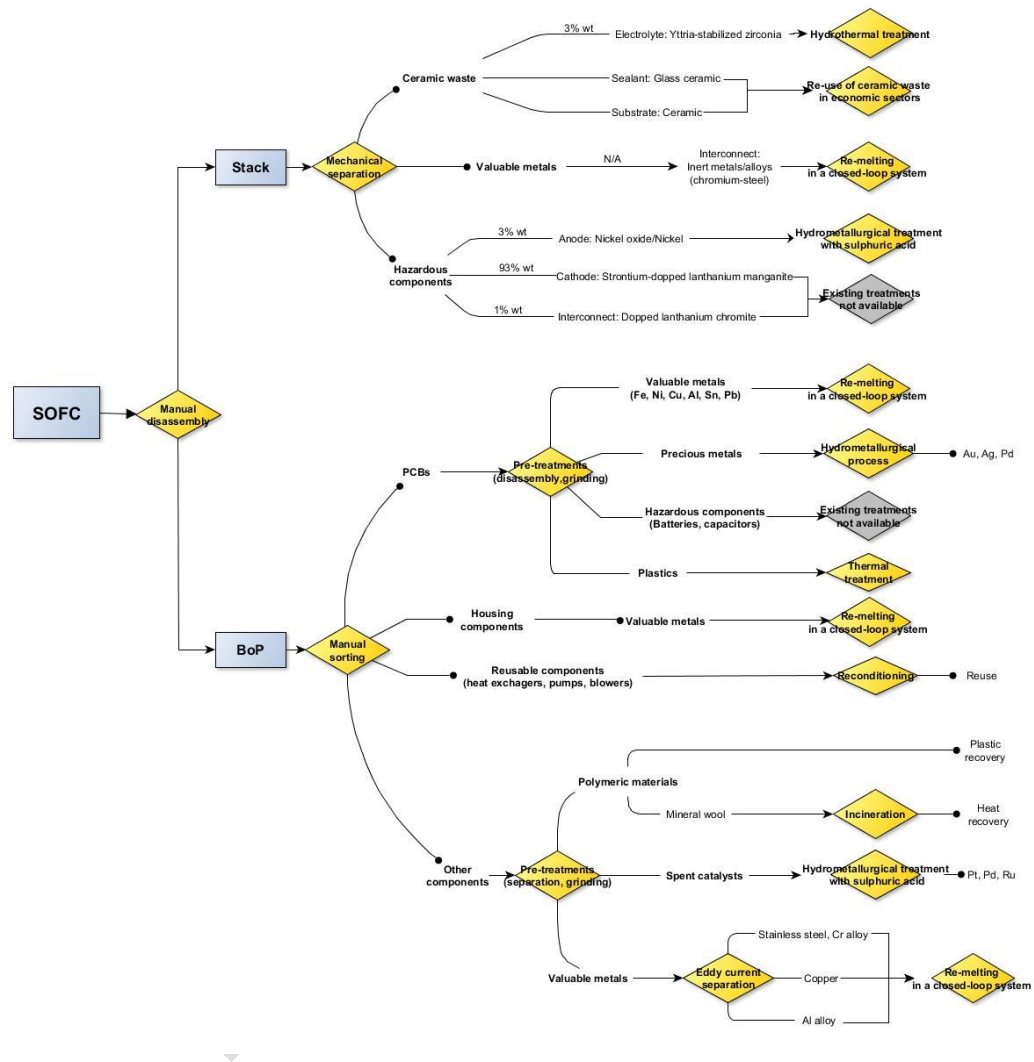
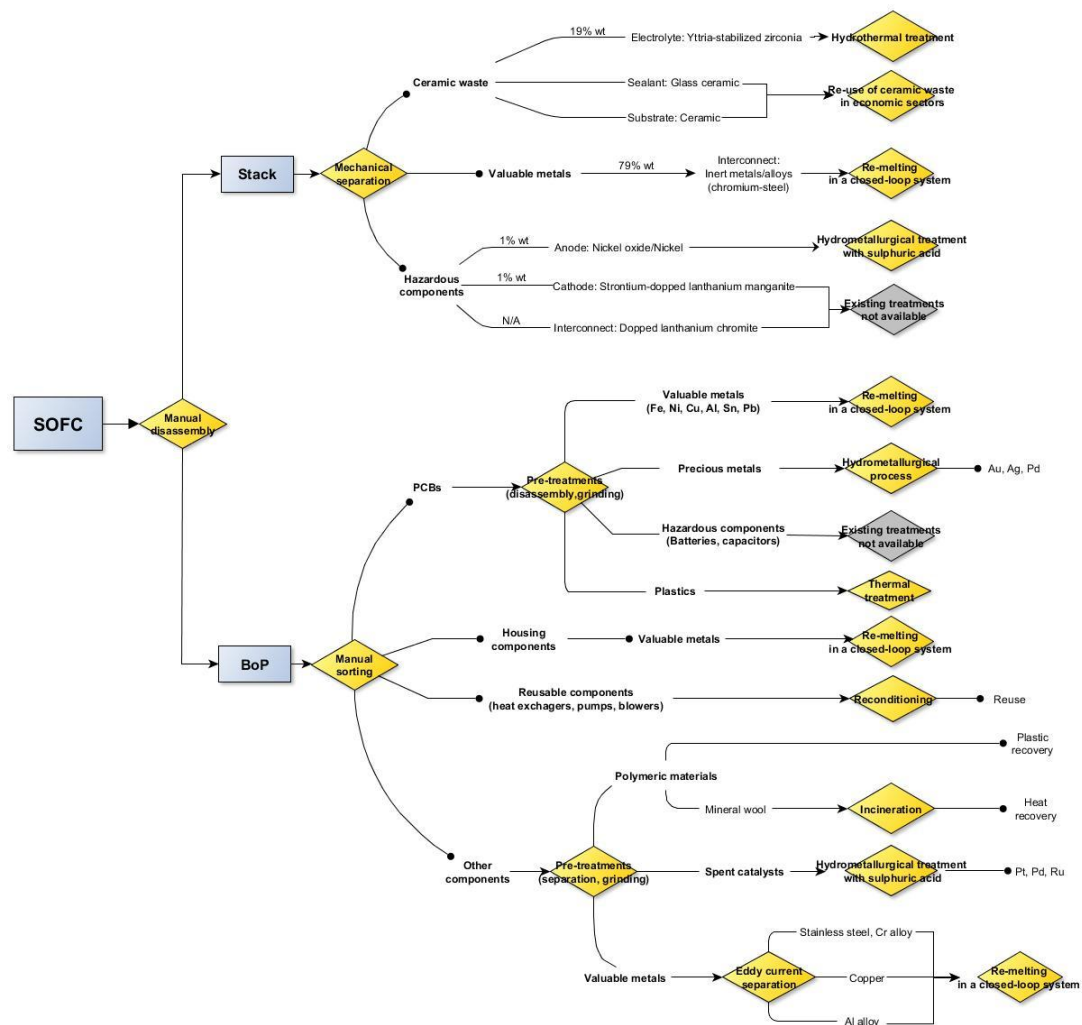**Figure 10. EoL scheme of SOFC tubular systems.**
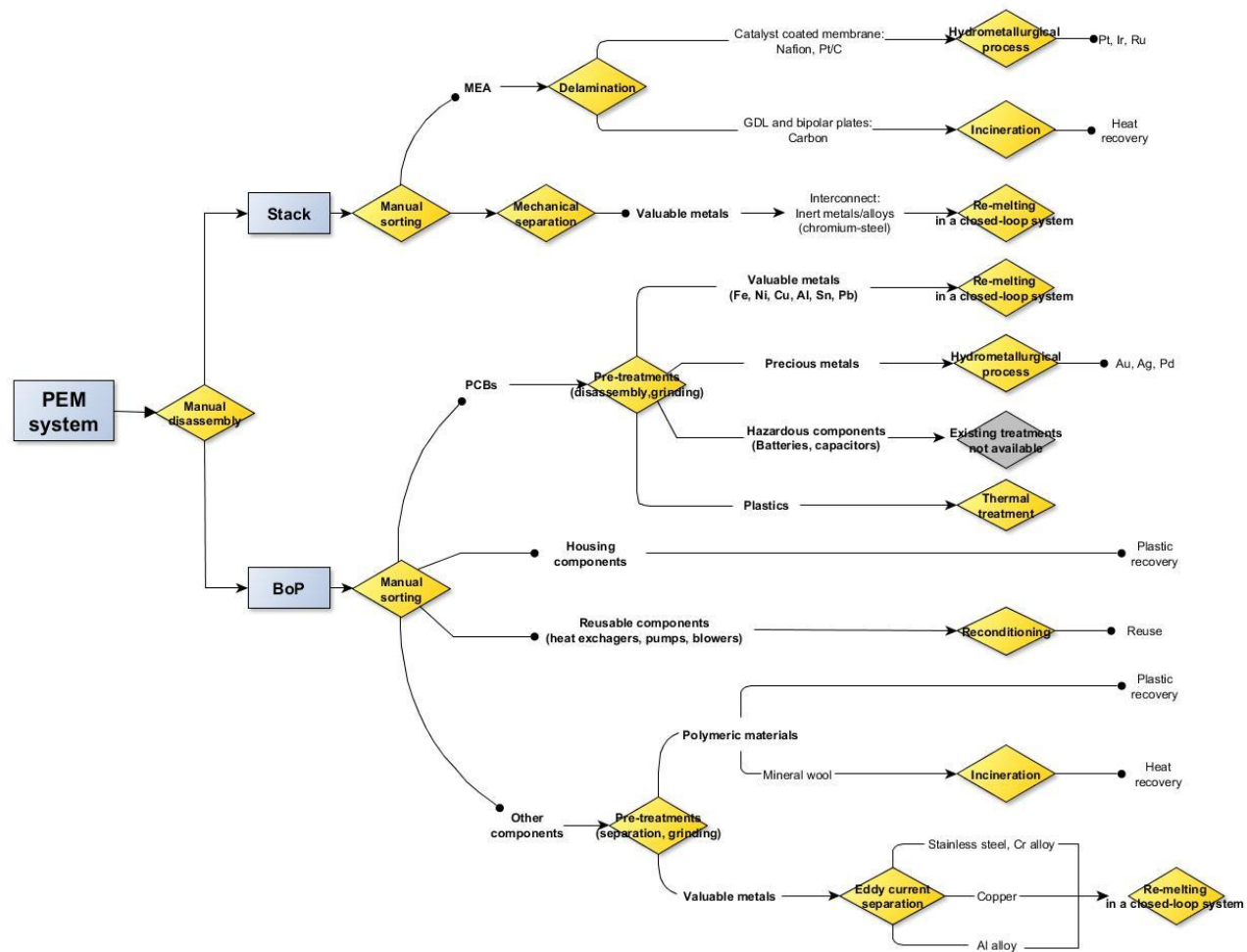
Figure 11. EoL scheme of SOFC planar systems.

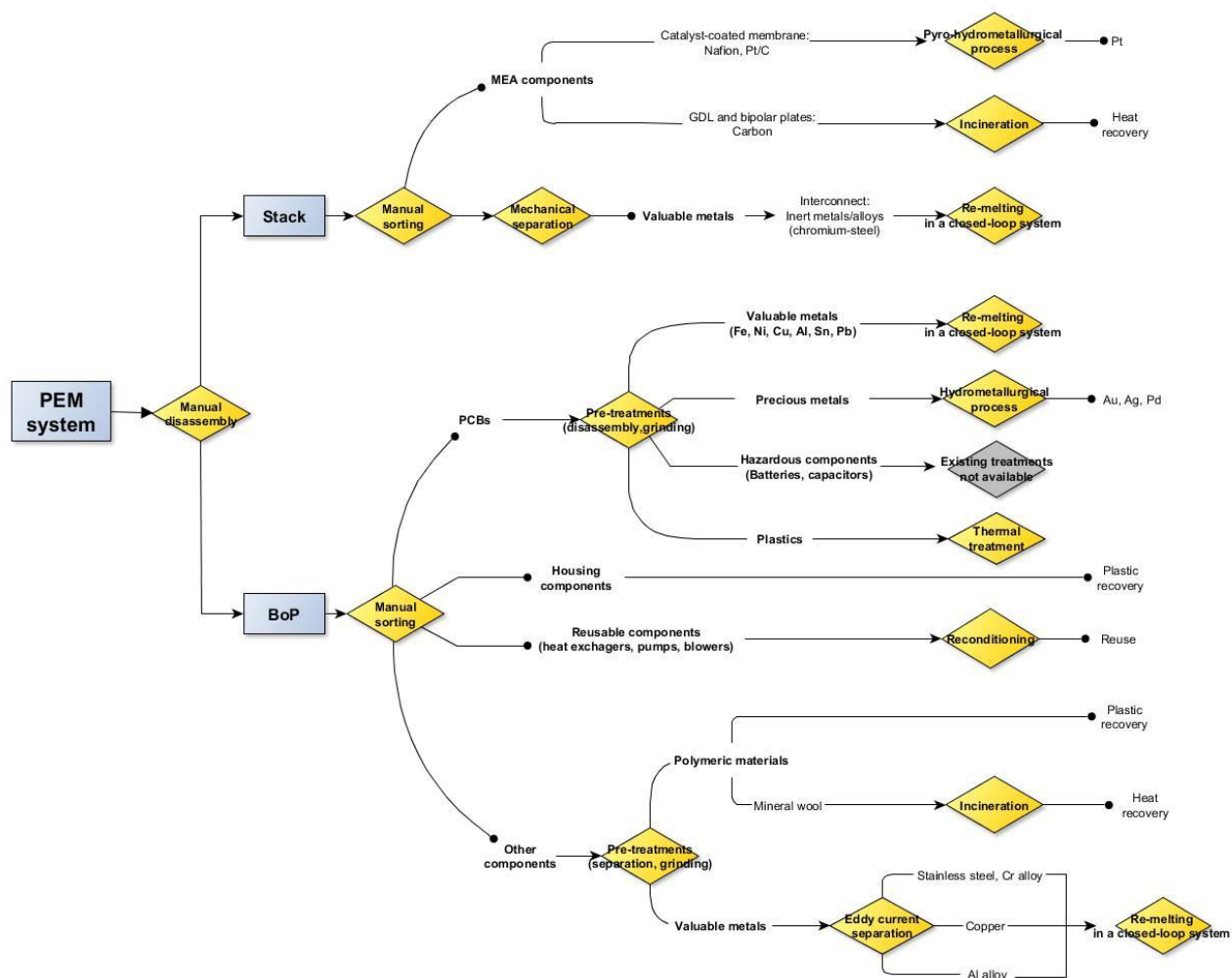Figure 12. EoL scheme of PEM systems: hydrometallurgical route.

Figure 13. EoL scheme of PEM systems: pyro-hydrometallurgical route.
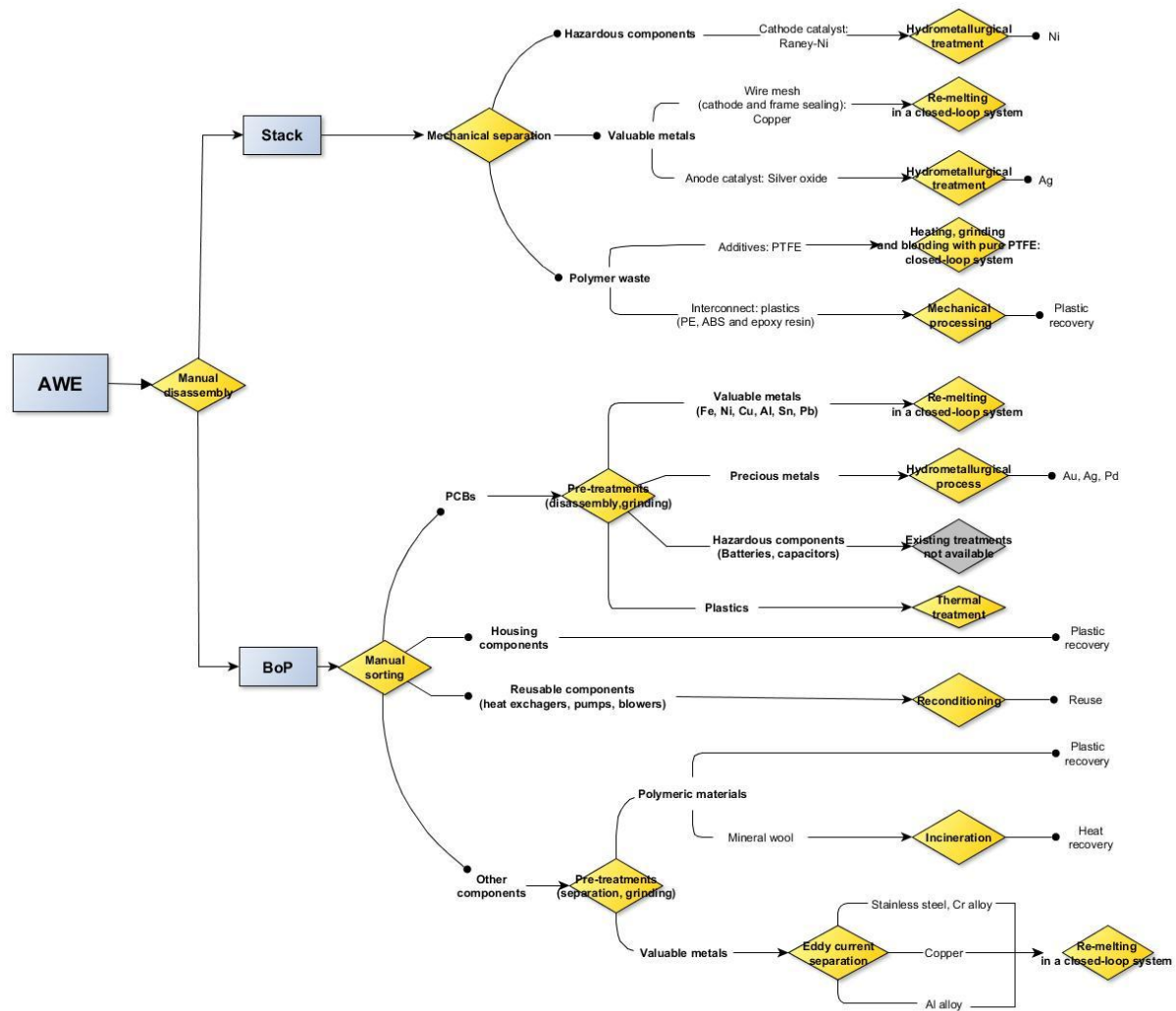
Figure 14. EoL scheme of AWE systems.

8. Conclusions

Currently, EoL strategies for PEMFCs, PEMWEs, AWEs and SOFCs are focused mainly on hydrometallurgical and pyrometallurgical recovery of precious metals used in the stacks as catalysts for the conversion process. Regarding BoP components, the literature also considers further recovery of valuable materials, such as metals and polymers for which mature recycling technologies are available. To face effectively the challenge of cost-competitiveness for the establishment of hydrogen economy, a full EoL strategy to reduce the costs of FCH devices is needed. However, there is a current lack of complete ecodesign and EoL strategies addressing exhaustively these FCH devices, which should include the recycling of valuable materials as well as reuse (or alternative use) options. The HyTechCycling project goes forward in this direction in order to fill this gap, and the findings of this document constitute the basis towards this target.

References

[1] United Nations Framework Convention on Climate Change, «Adoption of the Paris Agreement,» *UNFCCC*, Paris, 2015.

[2] L. Grandell, A. Lehtilä, M. Kivinen, T. Koljonen, S. Kihlman, L. S. Lauri, «Role of critical metals in the future markets of clean energy technologies,» *Renewable Energy*, vol. 95, pp. 53–62, 2016.

[3] D. Iribarren, M. Martín-Gamboa, J. Manzano, J. Dufour, «Assessing the social acceptance of hydrogen for transportation in Spain: An unintentional focus on target population for a potential hydrogen economy,» *International Journal of Hydrogen Energy*, vol. 41, pp. 5203–5208, 2016.

[4] E. I. Wright, A. J. Clegg, S. Rahimifard, L. Jones, «An investigation into end-of-life management of solid oxide fuel cells,» *3rd International Conference on Life Cycle Management*, University of Zurich, Zurich, 2007.

[5] J. T. S. Irvine, P. Connor, «Solid oxide fuels Cells: Facts and figures,» *Springer,* London, 2013.

[6] M. Gasik, editor, «Materials for fuel cells,» *Woodhead Publishing*, Cambridge, 2008.

[7] E. I. Wright, S. Rahimifard, «Strategic decision making for end-of-life management of fuel Ccells,» in *Leveraging Technology for a Sustainable World*, pp. 185–190, 2012.

[8] A. Brouzgou, S. Q. Song, P. Tsiakaras, «Low and non-platinum electrocatalysts for PEMFCs: Current status, challenges and prospects,» *Applied Catalysis B: Environmental*, vol. 127, pp. 371–388, 2012.

[9] O. Gröger, H. A. Gasteiger, J. P. Suchsland, «Review—Electromobility: Batteries or fuel cells?,» *Journal of the Electrochemical Society*, vol. 162, pp. A2605–A2622, 2015.

[10] S. Park, J. W. Lee, B. N. Popov, «A review of gas diffusion layer in PEM fuel cells: Materials and designs,» *International Journal of Hydrogen Energy*, vol. 37, pp. 5850–5865, 2012.

[11] X. Z. Yuan, H. Wang, J. Zhang, D. P. Wilkinson, «Bipolar plates for PEM fuel cells - From materials to processing,» *Journal of New Materials for Electrochemical Systems*, vol. 8, pp. 257–267, 2005.

[12] S. Asghari, M. H. Shahsamandi, M. R. Ashraf Khorasani, «Design and manufacturing of end plates of a 5 kW PEM fuel cell,» *International Journal of Hydrogen Energy*, vol. 35, pp. 9291–9297, 2010.

[13] R. Bhandari, A. C. Trudewind, P. Zapp, «Life cycle assessment of hydrogen production via electrolysis – A review,» *Forschungszentrum Jülich, Institute of Energy and Climate Research - Systems Analysis and Technology Evaluation (IEK-STE)*, Jülich, 2013.

[14] K. Zeng, D. Zhang, «Recent progress in alkaline water electrolysis for hydrogen production and applications,» *Progress in Energy and Combustion Science,* vol. 36, pp. 307–326, 2010.

[15] M. Bodner, A. Hofer, V. Hacker, «H_2 generation from alkaline electrolyzer,» *WIREs Energy and Environment,* vol. 4, pp. 365–381, 2015.

 [16] J. Hopewell, R. Dvorak, E. Kosior, «Plastics recycling: Challenges and opportunities,» *Philosophical Transactions of The Royal Society B Biological Sciences*, vol. 364, pp. 2115–2126, 2009.

[17] S. Papasavva, A. Coyle, S. Goldman, R. Privette, R. Legati, C. Huff, et al., «Development of recycling guidelines for PEM fuel cell systems,» *SAE Technical Paper 2003-01-1141,* 2003.

[18] Society of Automotive Engineers, «Recommended practice to design for recycling proton exchange membrane (PEM) fuel cell systems,» *SAE J 2594*, 2016.

[19] C. Hartnig, C. Roth, editors, «Polymer electrolyte membrane and direct methanol fuel cell technology – Volume 1,» *Woodhead Publishing*, Cambridge, 2012.

[20] M. Marafi, A. Stanislaus, «Spent catalyst waste management: A review. Part I – Developments in hydroprocessing catalyst waste reduction and use,» *Resources, Conservation and Recycling*, vol. 52, pp. 859–873, 2008.

[21] B. Singh, «Treatment of spent catalyst from the nitrogenous fertilizer industry – A review of the available methods of regeneration, recovery and disposal,» *Journal of Hazardous Materials,* vol. 167, pp. 24–37, 2009.

[22] S. Chatterjee, K. Kumar, «Effective electronic waste management and recycling process involving formal and non-formal sectors,» *International Journal of Physical Sciences*, vol. 4, pp. 893–905, 2009.

[23] F. Cucchiella, I. D'Adamo, S. C. Lenny Koh, P. Rosa, «A profitability assessment of European recycling processes treating printed circuit boards from waste electrical and electronic equipments,» *Renewable and Sustainable Energy Reviews*, vol. 64, pp. 749–760, 2016.

[24] M. Gurung, B. B. Adhikari, H. Kawakita, K. Ohto, K. Inoue, S. Alam, «Selective recovery of precious metals from acidic leach liquor of circuit boards of spent mobile phones using chemically modified persimmon tannin gel,» *Industrial & Engineering Chemistry Research*, vol. 51, pp. 11901–11913, 2012.

[25] L. Rocchetti, F. Vegliò, B. Kopacek, F. Beolchini, «Environmental impact assessment of hydrometallurgical processes for metal recovery from WEEE residues using a portable prototype plant,» *Environmental Science & Technology*, vol. 47, pp. 1581–1588, 2013.

[26] L. Zhang, Z. Xu, «A review of current progress of recycling technologies for metals from waste electrical and electronic equipment,» *Journal of Cleaner Production,* vol. 127, pp. 19–36, 2016.

[27] V. Goodship, A. Stevels editors, «Waste electrical and electronic equipment (WEEE) handbook,» *Woodhead Publishing*, Cambridge, 2012.

[28] B. D. James, J. M. Moton, W. G. Colella, «Mass production cost estimation of direct H_2 PEM fuel cell systems for transportation applications: 2014 update,» *ASME 2014 8th International Conference on Energy Sustainability*, 2014.

[29] L. Duclos, L. Svecova, V. Laforest, G. Mandil, P. X. Thivel, «Process development and optimization for platinum recovery from PEM fuel cell catalyst,» *Hydrometallurgy*, vol. 160, pp. 79–89, 2016.

[30] M. A. Barakat, M. H. H. Mahmoud, «Recovery of platinum from spent catalyst,» *Hydrometallurgy,* vol. 72, pp. 179–184, 2004.

[31] M. K. Jha, J. Lee, M. Kim, J. Jeong, B. S. Kim, V. Kumar, «Hydrometallurgical recovery/recycling of platinum by the leaching of spent catalysts: A review,» *Hydrometallurgy,* vol. 133, pp. 22–32, 2013.

[32] J. Y. Lee, S. V. Rao, B. N. Kumar, D. J. Kang, B. R. Reddy, «Nickel recovery from spent Raneynickel catalyst through dilute sulfuric acid leaching and soda ash precipitation,» *Journal of Hazardous Materials*, vol. 176, pp. 1122–1125, 2010.

[33] V. Coman, B. Robotin, P. Ilea, «Nickel recovery/removal from industrial wastes: A review,» *Resources, Conservation and Recycling,* vol. 73, pp. 229–238, 2013.

[34] N. Sathaiyan, V. Nandakumar, P. Ramachandran, «Hydrometallurgical recovery of silver from waste silver oxide button cells,» *Jorunal of Power Sources,* vol. 16, pp. 1463–1468, 2006.

[35] J. Zhao, X. He, J. Tian, C. Wan, C. Jiang, «Reclaim/recycle of Pt/C catalysts for PEMFC,» Energy *Conversion and Management*, vol. 48, pp. 450–543, 2007.

[36] T. Kojima, Y. Mori, M. Kamiya, R. Sasai, H. Itoh, «Disintegration process of yttria-stabilized zirconia ceramics using hydrothermal conditions,» *Journal of Materials Science,* vol. 42, pp. 6056–6061, 2007.

[37] C. Medina, M. I. Guerra, J. M. Morán, A. Juan, «Use of ceramic wastes in structurals concretes,» *1st Spanish National Conference on Advances in Materials Recycling and Eco – Energy*, Madrid, 2009.

[38] F. Puertas, I. García-Díaz, M. Palacios, M. F. Gazulla, M. P. Gómez, M. Orduña, «Clinkers and cements obtained from raw mix containing ceramic waste as a raw material. Characterization, hydration and leaching studies,» *Cement and Concrete Composites,* vol. 32, pp. 175–186, 2010.