

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

# Grant No. 700190

WP3 New strategies and technologies

# D3.1 New end-of-life technologies applicable to FCH products

Status: F

(D: Draft, FD: Final Draft, F: Final)

Dissemination level: PU

(PU: Public, CO: Confidential, only for members of the consortium (including the Commission Services))





This project has received funding from the Fuel Cells and Hydrogen 2 Joint Undertaking under grant agreement No 700190. This Joint Undertaking (JU) receives support from the European Union's Horizon 2020 research and innovation programme and Spain, Italy, Slovenia.

The contents of this document are provided "AS IS". It reflects only the authors' view and the JU is not responsible for any use that may be made of the information it contains.



# Document Change Control

Version Number	Date of issue	Author(s)	Brief description of changes
D3.1-v1	28 Nov 2017	Antonio Valente Diego Iribarren Javier Dufour (IMDEA Energy)	Complete version for review by partners.
D3.1-v2	20 Dec 2017	Antonio Valente Diego Iribarren Javier Dufour (IMDEA Energy)	Comments addressed. Final version.
D3.1-v3	13 Mar 2018	Antonio Valente Diego Iribarren Javier Dufour (IMDEA Energy)	Comments from EC FCH JU addressed. Final version.



### **Executive Summary**

This report constitutes the Deliverable 3.1 on new end-of-life (EoL) technologies applicable to fuel cells and hydrogen (FCH) products, which is associated with Task 3.1 "New proposed recycling and dismantling technologies applied to FCH technologies" within Work Package 3 "New strategies and technologies" of the HyTechCycling project. Different types of novel methods are searched in the literature and found to be applicable to the key FCH products (*viz.*, proton exchange membrane fuel cells/water electrolysers, alkaline water electrolysers, and solid oxide fuel cells) for the recovery of key materials in more efficient, safer and potentially cheaper ways than conventional existing EoL technologies. Novel technologies are found to be mainly applicable to the recovery of precious metals used in the stacks as catalysts. Regarding balance-of-plant (BoP) components, novel technologies focus on the recovery of valuable materials from printed circuit boards (PCBs).

The identification of the novel EoL technologies addressed in this deliverable, combined with the existing ones previously addressed in the Deliverable 2.2, leads to provide an overview of technology-oriented EoL strategies applicable to FCH systems for the recovery of critical materials. Techno-economic, regulatory and environmental aspects are contextualised and linked to the relevant EoL technologies through an analysis of potential strengths, weaknesses, opportunities, and threats (SWOT analysis).

Overall, existing and novel EoL technologies are not enough to define a full EoL strategy that reduces the costs of FCH devices and facilitates a well-established hydrogen economy. The HyTechCycling project goes forward in this direction.



## Contents

Document Change Control 3				
Executive Summary4				
Contents 5				
List of Figures				
List of Tables				
Abbreviations				
1. Introduction				
2. Structure of the work 10				
3. Novel EoL technologies applicable to FCH devices 11				
4. SOFC technology				
4.1. Novel technologies applicable to BoP components14				
5. PEMFC technology				
5.1. Selective electrochemical dissolution: Pt and C support recovery19				
5.2. Transient dissolution through potential alteration: Pt recovery				
5.3. Acid process: Nafion membrane and Pt recovery				
5.4. Alcohol solvent process: Nafion membrane and Pt recovery				
6. PEMWE technology				
7. AWE technology				
8. Strengths, Weaknesses, Opportunities, and Threats (SWOT)				
9. Conclusions				
References 29				



# List of Figures

Figure 1: Recovery processes applicable to FCH technologies: legend for the subsequent diagrams of Eo strategies at the FCH-technology level	L 2
Figure 2: Overall EoL strategy at the technology level for SOFCs 1	3
Figure 3: Weight and value distribution of materials in PCBs (based on [6])	5
Figure 4: General scheme for the recovery of PCB materials1	5
Figure 5: Material recovery from PCBs through supercritical methanol (based on [7])	6
Figure 6: Material recovery from PCBs through electrochemical technology (based on [9]) 1	7
Figure 7: Overall EoL strategy at the technology level for PEMFCs	8
Figure 8: Selective electrochemical dissolution (SED) of CCM based on [10]1	9
Figure 9: Platinum transient dissolution (TD) based on [11] 2	0
Figure 10: Nafion and catalyst recovery from PEM stacks through the novel acid process (AP) proposed in [12	2] 1
Figure 11: Alcohol solvent process (AD) based on patent US008124261B2 [13] 2	2
Figure 12: Overall EoL strategy at the technology level for PEMWEs 2	4
Figure 13: Overall EoL strategy at the technology level for AWEs 2	5
Figure 14: SWOT diagram for novel and existing EoL technologies applicable to FCH stacks 2	7



## List of Tables

Table 1: List of critical materials for FCH stacks and possible recovery techniques	. 11
Table 2: Conventional and novel EoL technologies applicable to FCH stacks	. 23



### **Abbreviations**

AD	Alcohol dissolution			
AP	Acid process			
AWE	Alkaline water electrolyser			
BMT	Biometallurgical technology			
BoP	Balance of plant			
ССВ	Catalyst-coated backing			
CCM	Catalyst-coated membrane			
EC	Electrochemical technology			
EoL	End of life			
FCH	Fuel cells and hydrogen			
GDL	Gas diffusion layer			
HDT	Hydrothermal technology			
HMME	Hydrometallurgical mild extraction technology			
HMT	Hydrometallurgical technology			
LSC	Doped lanthanum chromate (La <sub>0.85</sub> Sr <sub>0.15</sub> CrO <sub>3</sub> )			
LSM	Strontium-doped lanthanum manganese oxide (La <sub>0.85</sub> Sr <sub>0.15</sub> MnO <sub>3</sub> )			
MEA	Membrane electrode assembly			
PCB	Printed circuit board			
PEM	Proton exchange membrane			
PEMFC	Proton exchange membrane fuel cell			
PEMWE	Proton exchange membrane water electrolyser			
PFSA	Perfluorosulphonic acid polymer			
PGM	Platinum-group metals			
PMT	Pyrometallurgical technology			
PTFE	Polytetrafluoroethylene			
SCT	Supercritical technology			
SED	Selective electrochemical dissolution			
SOFC	Solid oxide fuel cell			
WEEE	Waste electrical and electronic equipment			
YSZ	Yttria-stabilised zirconia (ZrO <sub>2</sub> ) <sub>0.92</sub> (Y <sub>2</sub> O <sub>3</sub> ) <sub>0.08</sub>			



### 1. Introduction

The increasing energy demand and the awareness of the unsustainability of the current energy context are leading to a growing interest in renewable and clean energy alternatives. In this sense, hydrogen is expected to play a crucial role to match the requirements of a low-carbon society [1]. However, the deployment of hydrogen technologies, in terms of both production and use, is unavoidably affected by the feasibility of hydrogen devices.

To date, one of the most challenging barriers to overcome by hydrogen energy systems is the reduction of the costs associated with fuel cells and hydrogen (FCH) devices while maintaining their functional quality. In particular, the use of critical materials (especially as electro-catalysts) in the stack of these devices compromises their economic feasibility. In this regard, many authors have carried out experimental studies to identify potential alternative materials or attain improvements in efficiency and yield. Moreover, the low availability of critical materials (*e.g.*, Pt, Ru, and Ir) and their increasing global demand rise concerns about possible bottlenecks in the supply chains of FCH products.

Within this context, it is necessary to pursue end-of-life (EoL) strategies that optimise the recovery of the critical materials associated with FCH technologies. As a complement to previous studies on conventional existing technologies for the EoL of FCH devices [2], this document aims to identify and describe novel recovery technologies, showing how they can be applied to the EoL phase of FCH products in order to optimise their techno-environmental performance.



### 2. Structure of the work

A thorough literature review on novel EoL technologies applicable to FCH products is carried out in this report. A distinction is made between EoL technologies for stack components and those for balance-of-plant (BoP) components. It should be noted that the HyTechCycling target technologies are: proton exchange membrane fuel cells (PEMFC), proton exchange membrane water electrolysers (PEMWE), alkaline water electrolysers (AWE), and solid oxide fuel cells (SOFC).

The identification of novel EoL technologies and their combination with conventional existing ones addressed in previous tasks of the HyTechCycling project lead to present overall strategies applicable at the technology level for each FCH product under study. Broader strategies beyond the technology level are out of the scope of this report, but also covered within the framework of the HyTechCycling project (D3.2).



### 3. Novel EoL technologies applicable to FCH devices

With the exception of lanthanum compounds (for which no detailed information about their recovery has been found through the literature survey), a number of technologies are currently available to recover almost all critical materials present in FCH devices [2]. However, significant improvements are still needed regarding environmental and safety aspects, energy consumption, recovery efficiency, potential recovery of more than one product, *etc.* In other words, novel EoL technologies applicable to FCH products are needed. Furthermore, regarding BoP components, printed circuit boards (PCBs) may be present, mainly in the system control unit. PCBs are considered to be of special interest due to the presence of valuable metals (Au, Ag, Pd, and Cu; economic perspective) and flame-retardants containing bromine (their combustion releases furan and dioxins; environmental standpoint). Within this context, novel technologies applicable to critical stack materials and PCBs are reviewed hereinafter. At the technology level, the resulting EoL strategies are separately presented for each FCH product in Sections 4 -7.

Table 1 summarises existing and novel recovery technologies applicable to critical materials of FCH stacks. Metals belonging to the platinum group (Pt, Ru, Ir), whose reserves are depleting and which are associated with high economic costs, are the materials for which most of the EoL technologies have been found. In contrast, information as detailed as for PGMs is not available for novel EoL technologies applicable to rare earth elements (lanthanum compounds and YSZ) and nickel-based materials.

Device	Component	Material	Critical aspect	Recovery technologies		
			Cilical aspect	Existing <sup>a</sup>	Novel <sup>b</sup>	
	Anada	YZS	Cost; supply risk	HDT	N/A	
	Anoue	Ni; NiO	Hazard	HDT; HMT	N/A	
SOFC	Cathode	LSM	Hazard; supply risk	N/A	N/A	
SOLC	Electrolyte	YZS	Cost; supply risk	HDT	N/A	
	Intorconnocto	Ni; NiO	Hazard	HDT; HMT	N/A	
	Interconnects	LSC	Hazard; supply risk	N/A	N/A	
PEMFC	Anode	Pt	Cost	HMT; PMT	SED; TD; AP	
	Cathode	Pt	Cost	HMT; PMT	SED; TD; AP	
	Electrolyte	lonomer	Cost; hazard c	N/A	AD; AP	
PEMWE	Anode	lr; Ru	Cost; hazard	HMT; PMT	TD	
	Cathode	Pt	Cost	HMT; PMT	SED; TD; AP	
	Electrolyte	lonomer	Cost; hazard <sup>c</sup>	N/A	AD; AP	
	Bipolar plates	Ti	Cost	HMT	N/A	
AWE	Anode	Ag	Cost	HMT	N/A	
	Cathode	Ni; NiO	Hazard	HDT; HMT	N/A	

Table 1. I	lict of critical	matorials for	r ECU	ctacks and	noscible recover	tochniques
I dule I. L	LISE OF CITUCAL	IIIalei lais IU	ГГСП	SIDCKS dilu	possible recover	y lechingues

a HDT: hydrothermal technology; HMT: hydrometallurgical technology; PMT: pyrometallurgical technology

<sup>b</sup> TD: transient dissolution; AP: acid process; SED: selective electrochemical dissolution; AD: alcohol dissolution

<sup>c</sup> Concerns linked to HF emissions if the membrane is incinerated



#### D3.1 New end-of-life technologies applicable to FCH products

The EoL technologies applicable to FCH systems for the recovery of critical materials are summarised in Figure 1. Furthermore, the overall EoL strategies presented –at the technology level– in [2] for FCH products are updated herein by including novel technologies. The EoL strategies at the FCH-technology level are designed taking into account the waste management hierarchy defined in the European Union's Waste Framework Directive. The directive aims to maximise prevention and the reuse of devices, components and materials (recycling), while minimising landfill disposal and incineration. Therefore, for each of the four FCH products under study (Sections 4-7), after their collection, disassembly and material sorting, the configuration of the reprocessing activities (size reduction, and mechanical/chemical separation) is set to fulfil as far as possible the waste management hierarchy [3].



Figure 1: Recovery processes applicable to FCH technologies: legend for the subsequent diagrams of EoL strategies at the FCH-technology level



### 4. SOFC technology

Figure 2 shows the overall set of EoL management solutions applicable to SOFCs based on the components and materials involved. In SOFC systems, novel EoL technologies refer to the recovery of noble metals contained in PCBs within the system control unit (balance of plant), as detailed in Section 4.1.

Regarding stack components, no novel technologies are found to be applicable for the recovery of critical materials. In contrast, existing EoL technologies (hydrothermal and hydrometallurgical) are applicable for nickel recovery from the anode and interconnects as well as for the recovery of YSZ from spent electrolyte through the hydrothermal route. Nonetheless, due to the high operating temperature of SOFCs, high-purity materials are needed to avoid performance degradation. This means that the use of recovered YSZ is possible in open-loop recycling, *e.g.* for its application in electrical/electrochemical sectors with a lower grade than the material for the SOFC anode or electrolyte. Alternatively, if hydrothermal recovery is not applied, the ceramic composite material can –after grinding and mechanical separation– be recycled, still in an open-loop scheme, for use in construction applications.

Regarding lanthanum compounds (LSM and LSC), which can be present in interconnects and/or the cathode material, no recovery process is found to be available and, due to their hazardous nature, they are disposed in hazardous landfills.







#### 4.1. Novel technologies applicable to BoP components

The general strategy for the recovery of BoP ancillary components (blowers, pumps, compressors, heat exchangers, housing components, *etc.*) is focused on their re-conditioning for reusing them when manufacturing a new FCH system [2]. Other materials, such as plastics, ferrous and non-ferrous metals contained in wires, valves, pipes and other BoP components, are instead separated through well-established techniques involving size reduction followed by physical separation through gravimetric, magnetic and electrostatic techniques. Metals, in general, can be recovered with similar chemical and mechanical properties to those of the virgin material. After their recovery, they can be used in applications with a similar value to the original one (closed-loop recycling). Under these circumstances, the energy and environmental burdens of the virgin material are avoided. However, when the intrinsic properties of the recycled material are significantly worse than those of the virgin material, then its use is unavoidably linked to lower-value applications (open-loop recycling) [4].

PCBs present in the BoP of FCH devices, especially in the system control unit, are potential sources of secondary raw materials since they are rich in precious metals (*e.g.*, silver, gold, and palladium) and other valuable metals such as copper, tin, and nickel. Their concentration in the respective primary sources (ores) is significantly lower than the average concentration in PCBs (up to 25 times more concentrated in weight). Therefore, their recovery is associated with potential economic and environmental benefits thanks to the avoidance of the environmental impacts associated with their primary extraction [5].

Figure 3 shows the composition of PCBs estimated in [6] along with the economic share of each material. On the one hand, gold is the metal that dominates the economic profile of PCBs. Copper, nickel, aluminium and silver also have a significant contribution in economic terms. Hence, the recovery of these metals should be addressed. On the other hand, the label "other" includes resins and glass fibres, which show negligible economic relevance even though they dominate the weight of PCBs. Another critical EoL aspect of PCBs refers to environmental issues due to incineration or landfilling. The presence of flame-retardants and the high concentration of lead (Pb) and polybrominated compounds make the correct recycling of PCBs essential.

The general scheme for the recovery of PCB materials is shown in Figure 4. Conventional treatments for a cost-competitive recovery of precious metals from PCBs are based on hydrometallurgical (HMT) or pyrometallurgical (PMT) processes [2]. Furthermore, novel techniques applicable to PCBs are under evaluation. In this regard, almost all metals in PCBs can be recovered in solid mixture state through supercritical technologies (SCT), or in solution through hydrometallurgical mild extracting technologies



(HMME). Moreover, a few specific metals can be recovered through biometallurgical (in solution; BMT), electrochemical (in solid mixture state; EC) and vacuum metallurgical (single metal in solid state; VM) technologies [6].



Figure 3: Weight and value distribution of materials in PCBs (based on [6])



Figure 4: General scheme for the recovery of PCB materials



Regarding HMME, working conditions are safe and environmentally friendly. However, the high costs of the reactants strongly affect the economic performance of this technique.

SCT options (*e.g.*, Figure 5) involve very high recovery efficiency and affordable capital costs. Moreover, they are environmentally friendly and compact. On the other hand, operating and maintenance costs can be relatively high because of the energy demand to reach supercritical conditions. These conditions mean also a disadvantage under safety aspects.



#### Figure 5: Material recovery from PCBs through supercritical methanol (based on [7])

BMT options are gaining interest due to outstanding economic and environmental performances [8]. However, these technologies need long periods for leaching, the metals need to be exposed on the surface layer, and the process is highly sensitive to working conditions such as temperature, pH, and the concentration of oxygen and nutrients.

EC options are mature and they need low costs of investment. Nevertheless, their level of industrialisation is still low, and they are not yet considered safe and environmentally friendly. Figure 6 shows a technique proposed by Kim *et al.* [9] for the recovery of copper, lead, tin and zinc contained in waste PCBs.

VM options have interesting techno-environmental features, including a high recovery rate of rare earth elements in WEEEs. They are highly-selective technologies, especially suitable for high vapour pressure metals. Nevertheless, they are still in a low level of industrialisation, and the technology needs to be further investigated to reach a better techno-economic performance.

Finally, other non-industrialised technologies, such as ultrasonic and mechano-chemical ones, are gaining the interest of researchers thanks to their low cost and good environmental profile. They can be combined with other technologies to improve the overall system performance.





Figure 6: Material recovery from PCBs through electrochemical technology (based on [9])



### 5. PEMFC technology

Figure 7 shows the overall EoL scheme applicable to PEMFC devices. In these devices, the physical and chemical structure of bipolar plate materials such as graphite, stainless steel and carbon composites is expected to be altered during operation. Hence, their reuse or recovery for the same original application is not possible. An opportunity for the materials recovered from bipolar plates is their use as insulation raw material for electronic devices or in steel manufacturing (open-loop recycling).

Regarding MEA components, existing technologies (hydrothermal and hydrometallurgical) and novel technologies (acid process, transient dissolution, and selective electrochemical dissolution; Sections 5.1-5.3) are found to be applicable for the recovery of PGM metals from the electrodes along with other valuable materials such as ionomers from the membrane or the carbon support of the noble catalyst. Alcohol dissolution (Section 5.4) is also applicable as a pre-treatment for the recovery of ionomers from the membrane before the catalyst recovery process. The recovery of MEA's critical materials allows recycling in a closed-loop scheme, which means a potentially higher benefit for the FCH sector with respect to open-loop recycling. Regarding BoP components, the novel technologies explained in Section 4.1 can also be applied to PEMFCs.



Figure 7: Overall EoL strategy at the technology level for PEMFCs



#### 5.1. Selective electrochemical dissolution: Pt and C support recovery

Latsuzbaia *et al.* [10] proposed a novel electrochemical process that allows recovering separately both a high percentage of catalyst and its carbon support (Figure 8). With different pH, voltage and temperature window, the method can be extended to other catalyst materials supported in other electrochemical devices. The main advantages are a high purity of the recovered catalyst, the mild conditions regarding pH, temperature and voltage, the good techno-environmental performance, and the fact that the electrodes can be easily recast *in situ*. As the corrosion of the carbon support occurs at potentials over 1.2 V, the method works at potentials below this value during the dissolution process, thus allowing the recovery of the carbon support. On the other hand, the process requires electricity (though less than conventional electrochemical processes), and the carbon support requires reconditioning before reuse.



Figure 8: Selective electrochemical dissolution (SED) of CCM based on [10]

#### 5.2. Transient dissolution through potential alteration: Pt recovery

Hodnik *et al.* [11] proposed a novel method based on the dissolution –in HCI media– of Pt from EoL materials (Figure 9). The method involves a cyclic change in the oxidation state of the Pt surface. During the transient phase, the dissolution of Pt takes place in a low-concentrated acid solution. O<sub>3</sub> and CO gases –oxidising and reducing agents, respectively– are used to cyclically change the oxidation state of the Pt surface, which avoids the use of external potential and electrodes. The mild working conditions, the high recovery yield and the avoided use of external potential represent the main advantages with respect to the method proposed by Latsuzbaia *et al.* [10]. The method proposed can be used for the recovery of other critical metals relevant to the HyTechCycling project, such as Pd, Ru, and Ir (in AWEs and PEMWEs). This would be done by adjusting specific parameters such as the concentration of the lixiviating agents and the number and duration of the cycles. On the other hand, the possibility of recovering other stack materials



such as the catalyst support or the membrane is not reported, and the duration of the process is highly influenced by the concentration of acid reactants and the operating temperature. In this respect, the higher the pH and the temperature, the shorter the time scale of the process.



Figure 9: Platinum transient dissolution (TD) based on [11]

### 5.3. Acid process: Nafion membrane and Pt recovery

Xu *et al.* [12] proposed a novel acid process configuration (Figure 10). This involves a promising approach that allows the efficient recovery of both the Nafion membrane and a high percentage of platinum from the CCM of PEM stacks by using strong acids to oxidise the carbon support. One of the main advantages of this process is the possibility of re-casting Nafion and recovering platinum for new stacks with potentially excellent electrochemical performance.

Besides the possibility of recovering Pt at a high yield, this process has the advantage of recycling PFSA, which constitutes the Nafion, to recast the polymeric membrane. When compared to conventional hydrometallurgical and pyrometallurgical processes, the problems associated with a lower yield of PGM recovery and with HF emissions, respectively, are avoided. On the other hand, the main disadvantages of the method are the relatively long chain and duration, and –as usual for acid recovery processes– the harsh conditions regarding pH.







Figure 10: Nafion and catalyst recovery from PEM stacks through the novel acid process (AP) proposed in [12]

#### 5.4. Alcohol solvent process: Nafion membrane and Pt recovery

A patent by Shore [13] addresses the recovery of noble metals and the Nafion membrane from PEM devices (both fuel cells and electrolysers) using an alcohol solvent process (Figure 11). This invention may be thought as a pre-treatment allowing the recovery of the catalyst as well as of the polymer resin, which can be re-converted into Nafion. The MEA (including GDLs and other layers) is delaminated under the action of solvents (alkyl alcohol solution) and a mixer/agitator in the delamination tank. The recycling of the solvent improves the profile of this process concerning its techno-environmental performance and cost reduction. The microwave heater increases the temperature of the solution up to the range suitable for the dissolution of the polymeric membrane. A critical parameter is the polymer particle size in the dispersion in order to allow the subsequent separation through a filter press and ultra-filtration. This parameter is controlled in the microwave heater by changing temperature and residence time. In a successive step, the solvent is separated from larger particles through a screen and pumped to a filter press. On the one hand, mild working conditions in terms of pH, temperature and voltage favour the safety aspects of this process. On the other hand, this technology represents an increase in the global complexity of the recovery process, which compromises its economic feasibility.





Figure 11: Alcohol solvent process (AD) based on patent US008124261B2 [13]

A similar invention has been presented by Stephen Grot and Walther Grot [14], who is the developer of Nafion for DuPont [15]. It allows separating the ionomer from MEA in a dissolved form through an autoclave reactor using an alcohol/water solution, and a subsequent stage of centrifugation at 15000 rpm. The two resulting streams are the Nafion solution and the Pt/C ink. While devices using recycled Nafion show good electrochemical performances, fuel cells using recycled Pt/C ink from this process show electrochemical performances significantly worse than those of the original device. This is due to the incomplete removal of the Pt from the discharge stream. In particular, the GDL mixing with the catalyst hinders complete separation. Therefore, the primary interest of this patent is the recovery of the ionomer that, though not as valuable as the Pt, has a relatively high value.

Finally, Table 2 summarises the main technical aspects of both conventional and novel EoL technologies applicable to FCH stack materials.



#### D3.1 New end-of-life technologies applicable to FCH products

Table 2: Conventional and novel EoL technologies applicable to FCH stacks								
Process <sup>a</sup>	Working	Develoption	Energy requirements		Catalyst	Further	Duralla	<u> </u>
	conditions	Reactants	Heat	Electricity	yield	materials	Duration	Comment
SED	Harsh (T)	HCIO <sub>4</sub> , O <sub>2</sub> , HCI	-	Moderate	~90%	C support	3-5 h	-
TD	Mild	NaCI, HCI, CO, O <sub>3</sub>	-	-	~90%	-	~3 h	-
AP	Harsh (pH)	H <sub>2</sub> SO <sub>4</sub> , NaOH, Cu	Moderate	High	> 94%	lonomer	>80 h	-
AD	Mild	Alcohol and water	-	Moderate	~90%	lonomer	< 1 h	Pre-treatment
PMT	Harsh (pH,T)	Acids, HCI, NaOH, HNO3	High	-	~90%	-	~10 h	Existing technology [2]
HMT	Harsh (pH)	Oxidants, HCI, NH4CI, NaOH	-	-	< 80%	-	>24 h	Existing technology [2]
HDT	Harsh (T)	Water	High	Moderate	-	-	~24 h	Existing technology [2]

<sup>a</sup> SED: selective electrochemical dissolution; TD: transient dissolution; AP: acid process; AD: alcohol dissolution; PMT: pyrometallurgical; HMT: hydrometallurgical; HDT: hydrothermal



### 6. PEMWE technology

Figure 12 shows the overall EoL scheme applicable to PEMWE devices. Similarly to PEMFC stacks, novel and existing EoL technologies are applicable to the critical materials of the device's MEA. The anode electrocatalyst for PEMWEs can be iridium or ruthenium, which are found to be recoverable through existing (in particular, pyrometallurgical and hydrometallurgical processes) or novel (transient dissolution) methods. As observed for PEMFC products, the application of the EoL technologies already described in Sections 4.1 and 5.1-5.4 would allow recycling BoP and stacks' materials in closed-loop schemes.

PEMWE's bipolar plates are conventionally made of titanium alloys. Generally, titanium can be recovered through conventional methods based on physical separation (size reduction and magnetic separation); however, being combined with other elements, its recovery requires more complex processes, *e.g.* hydrometallurgical processes. No novel recovery technologies are found for titanium recovery from PEMWE systems.



Figure 12: Overall EoL strategy at the technology level for PEMWEs



### 7. AWE technology

Figure 13 represents the overall EoL picture for AWE systems. At the AWE stack level, existing technologies are found to be applicable for the recovery of silver as the anode electrocatalyst and nickel compounds as the cathode electrocatalyst. The application of these methods would allow recycling Ni and Ag in a closed-loop scheme. No novel recovery methods are found to be applicable to these AWE materials. It is worth mentioning that asbestos –banned in the EU in 2005 [16]– used in older devices as the AWE diaphragm could also be recovered through thermal processing to produce harmless materials such as silicate glass or ceramic bricks. Regarding BoP components –as observed for SOFC, PEMFC and PEMWE devices–, novel EoL technologies can be applied for the recovery of valuable materials from PCBs (Section 4.1).



Figure 13: Overall EoL strategy at the technology level for AWEs



### 8. Strengths, Weaknesses, Opportunities, and Threats (SWOT)

Existing and emerging technologies applicable to FCH stacks are further evaluated in this section through a SWOT analysis (Figure 14). Thus, the analysis takes into account strengths, weaknesses, opportunities and threats identified within the technical, economic, environmental, social and regulatory dimensions.

Regarding economic aspects, PMT is associated with a relatively adverse profile. High operating and investment costs –along with threats of potential taxes due to the use of hazardous reactants and the high level of emissions– could negatively affect the economic performance of this type of technology when compared to the novel ones, even though economic benefits are feasible. AP shows similar weaknesses and, even though they are partially balanced by a better technical profile regarding both efficiency and number of recoverable materials, the high electricity demand leads to economic concerns regarding the potential future increase in electricity prices. In fact, the threat of high electricity prices affects the economic performance of all those processes with a high or moderate electricity demand.

In contrast, SED and TD processes show potential economic advantages because of low investment costs and high recovery efficiency along with high purity of the recovered metal. Deciding which one can be considered the best option in economic terms is intricate. On the one hand, even though SED allows the recovery not only of the catalyst but also of the carbon support, the use of electricity (although moderate) exposes this technology to the risk of an increase in electricity prices. On the other hand, TD makes use of toxic (CO) and oxidant (O<sub>3</sub>) reactants, thus exposing this technology to the risk of future regulatory restrictions or taxes on the use hazardous raw materials.

Regarding technical features, the strength "versatility" should be understood as the possibility of processing components that come not only from FCH devices but also from other sources and sectors, thereby mitigating the risk of abandonment of FCH technology if ruled out by policy-makers.



#### D3.1 New end-of-life technologies applicable to FCH products

#### STRENGTHS

PMT: S4, S10, S11, S12, S14 HMT: S1, S2, S7, S10, S11, S14 AP: S4, S5, S9, S10, S14 SED: S1, S4, S5, S6, S8, S14 AD: S2, S3, S4, S5, S6, S9, S13 TD: S1, S3, S4, S6, S10, S13, S14

#### WEAKNESSES

PMT: W1, W2, W3, W5, W7, W9, W10 HMT: W3, W4, W6, W9 AP: W1, W2, W3, W6, W7, W9, W10 SED: W7, W9 AD: W1, W7, W8 TD: W9

OPPORTUNITIES	THREATS
PMT: 04, 05	<b>PMT</b> : T1, T2
HMT: O4, O5	HMT: T1
AP: O2, O4, O5	AP: T1, T2, T4
SED: O2, O3, O4, O5	SED: T1, T3, T4
AD: O1, O2, O3, O4, O5	AD: T3, T4
TD: O1, O2, O4, O5	TD: T1

#### Strengths

- S1: Low investment cost
- S2: Low operating costs
- S3: Mild operating conditions
- S4: High recovery efficiency
- S5: Recovery of more than one material
- S6: Fast process
- S7: Low energy requirements
- S8: Low complexity
- S9: Toxic compound removal
- S10: Versatile technology for other sectors
- S11: Mature technology
- S12: Co-processing of material from different sources
- S13: Low environmental concerns
- S14: High potential for reuse in high-value application

#### Opportunities

- O1: Potential economic incentives for eco-friendly techniques
- O2: Potential breakthrough in technology
- O3: Anticipated fulfilment of future circular economy targets
- O4: High deployment of FCH technologies
- O5: High social demand of green market

#### Weaknesses

- W1: High investment costs
- W2: High operating costs
- W3: Harsh operating conditions
- W4: Low recovery efficiency
- W5: Applicable to only one type of material
- W6: Lengthy process
- W7: High energy requirements
- W8: Low-value product
- W9: Use of hazardous reactants
- W10 Significant environmental concerns

#### Threats

- T1: More severe regulations on hazardous materials
- T2: More severe restrictions on emission levels
- T3: FCH deployment ruled out by policy-makers
- T4: Increased electricity prices

Figure 14: SWOT diagram for novel and existing EoL technologies applicable to FCH stacks



### 9. Conclusions

Novel EoL technologies for FCH stack materials are mainly oriented towards the recovery of precious metals. When compared to conventional EoL technologies, the main advantages associated with the reviewed emerging technologies are related to the possibility of recovering more than one valuable product (ionomer or carbon support for PEM-based systems) in addition to the precious metals. Furthermore, novel technologies are usually linked to enhanced technical performances (*e.g.*, in terms of process duration), improved economic and environmental performances (thanks to the lower amount of energy required), and safer working conditions (thanks to mild operating conditions in terms of temperature, pH and voltage, as well as to the use of non-hazardous reactants).

Regarding BoP components, novel technologies are found to be applicable mainly to PCBs, which are rich in high-value metals and for which current recovery practices lead to significant environmental concerns. Although several emerging technologies could be applied, further research is still needed due to the low level of industrialisation. In general, the main trend for BoP components is oriented towards the reuse of components such as pumps, blowers, compressors, *etc.* 

When compared to conventional EoL technologies such as pyrometallurgical processes, TD generally shows a more favourable profile with relevant advantages in terms of versatility and economic performance. SED and AD could also be considered promising EoL technologies, but their application is highly conditioned by the actual commercial deployment of PEM devices. In this sense, and in order to effectively face the challenge of a well-established hydrogen economy, a full EoL strategy beyond the technology level is still required. The HyTechCycling project progresses in this direction.



#### References

- [1] International Energy Agency, Technology Roadmap, Paris, 2015.
- [2] A. Valente, M. Martín-Gamboa, D. Iribarren, J. Dufour, FCH JU HyTechCycling project D2.2 Existing end-of-life technologies applicable to FCH products, 2018. http://hytechcycling.eu/downloads/deliverables/.
- [3] European Parliament and Council, Directive 2008/98/EC of the European Parliament and of the Council of 19 November 2008 on waste and repealing certain directives, 2008.
- [4] S. Huysman, S. Debaveye, T. Schaubroeck, S. De Meester, F. Ardente, F. Mathieux, J. Dewulf, The recyclability benefit rate of closed-loop and open-loop systems: A case study on plastic recycling in Flanders, Resour. Conserv. Recycl. 101 (2015) 53–60. doi:10.1016/j.resconrec.2015.05.014.
- [5] H.M. Veit, A.M. Bernardes, A.C. Kasper, N.C. de F. Juchneski, C.O. Calgaro, E.H. Tanabe, Electronic Waste - Recycling Techniques, Springer, Dordrecht Heidelberg London New York, 2015. doi:10.1007/978-3-319-15714-6.
- [6] L. Zhang, Z. Xu, A Review of Current Progress of Recycling Technologies for Metals from Waste Electrical and Electronic Equipment, J. Clean. Prod. 127 (2016) 19–36. doi:10.1016/j.jclepro.2016.04.004.
- [7] F.R. Xiu, F.S. Zhang, Materials recovery from waste printed circuit boards by supercritical methanol, J. Hazard. Mater. 178 (2010) 628–634. doi:10.1016/j.jhazmat.2010.01.131.
- [8] S. Syed, Silver recovery aqueous techniques from diverse sources: Hydrometallurgy in recycling, Waste Manag. 50 (2016) 234–256. doi:10.1016/j.wasman.2016.02.006.
- [9] E.Y. Kim, M.S. Kim, J.C. Lee, J. Jeong, B.D. Pandey, Leaching kinetics of copper from waste printed circuit boards by electro-generated chlorine in HCl solution, Hydrometallurgy 107 (2011) 124–132. doi:10.1016/j.hydromet.2011.02.009.
- [10] R. Latsuzbaia, E. Negro, G.J.M. Koper, Environmentally friendly carbon-preserving recovery of noble metals from supported fuel cell catalysts, ChemSusChem 8 (2015) 1926–1934. doi:10.1002/cssc.201500019.
- [11] N. Hodnik, C. Baldizzone, G. Polymeros, S. Geiger, J.-P. Grote, S. Cherevko, A. Mingers, A. Zeradjanin, K.J.J. Mayrhofer, Platinum recycling going green via induced surface potential alteration enabling fast and efficient dissolution, Nat. Commun. 7 (2016) 13164. doi:10.1038/ncomms13164.
- [12] F. Xu, S. Mu, M. Pan, Recycling of membrane electrode assembly of PEMFC by acid processing, Int. J. Hydrogen Energy. 35 (2010) 2976–2979. doi:10.1016/j.ijhydene.2009.05.087.
- [13] L. Shore, Process for Recycling Components PEM Fuel Cell Membrane Electrode Assembly, US008124261B2, 2012.
- [14] S. Grot, W. Grot, Recycling of used perfluorosulfonic acid membrane, US 7255798 B2, 2007.
- [15] DuPont, Official web site, (2018). http://www.dupont.com (accessed January 8, 2018).
- [16] European Chemical Agency, Annex XV Restriction Report Amendment to a Restriction Substance Name: Chrysotile IUPAC Name: Chrysotile EC Number: - CAS Number(S): CAS No 12001-29-5 and 132207-32-0, 2014.