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 3.2 on new strategies for technologies applicable to fuel cells and hydrogen (FCH) products, which is associated with Task 3.2 “New strategies for FCH technologies in the phase of recycling and dismantling” within Work Package 3 “New strategies and technologies” of the HyTechCycling project. The main purpose of this document is to deliver a reference document paving the way for a more comprehensive guide to enhancing the supply chain of FCH technologies. The key aim is to provide valuable information, since the design phase, to help manufacturers to optimise the life-cycle stages of FCH technologies in terms of cost-effectiveness and environmental concerns linked to emissions, use of critical materials, and waste management. In order to efficiently face the challenge of cost-competitiveness for a well-established hydrogen economy, a full end-of-life (EoL) strategy to reduce the criticalities of FCH devices is required.

Relevant EoL strategies are proposed addressing the main actors identified in the supply chain, and considering design, manufacture and logistics of the FCH products under evaluation. The strategies are defined considering the role and the operation performed by raw material suppliers, FCH components' suppliers, FCH manufacturers, FCH users, and waste managers. The role of a specialised recovery centre is emphasised in different scenarios of FCH market deployment.

Strategies applicable to the design stage are also highlighted through an extensive literature survey focused on the identification of manufacturing and designing techniques that lead to the reduction or replacement of critical materials for each of the selected FCH products.

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Abbreviations

AWE	Alkaline water electrolyser
BoP	Balance of plant
CCM	Catalyst-coated membrane
EoL	End of life
FCH	Fuel cell and hydrogen
GDL	Gas diffusion layer
LCA	Life Cycle Assessment
LSM	Strontium-doped lanthanum manganese oxide ($La_{0.85}Sr_{0.15}MnO_3$)
LSC	Doped lanthanum chromate ($La_{0.85}Sr_{0.15}CrO_3$)
MEA	Membrane electrode assembly
ORR	Oxygen reduction reaction
PEM	Proton exchange membrane
PEMFC	Proton exchange membrane fuel cell
PEMWE	Proton exchange membrane water electrolyser
PFSA	Perfluorosulfonic acid polymer
PGM	Platinum-group metals
PTFE	Polytetrafluoroethylene
RC	Recovery centre
REACH	Registration, Evaluation, Authorisation and Restriction of Chemicals
RoHS	Restriction of the use of certain Hazardous Substances
SOFC	Solid oxide fuel cell
YSZ	Yttria-stabilised zirconia $(ZrO_2)_{0.92}(Y_2O_3)_{0.08}$
WEEE	Waste electrical and electronic equipment

1. Introduction

We are currently in the middle of an energy transition, moving from fossil to sustainable fuels. The European Union is undertaking policies and strategies that prioritise making energy more secure, affordable and sustainable [1]. The EU targets for 2020 aim to cut greenhouse gas emissions by 20% with respect to 1990, to have 20% renewable share in energy consumption, and to improve efficiency by 20% (20-20-20 targets); these targets become 40%, 27% and 27%, respectively, with 2030 horizon. The use of FCH products (*viz.*, PEMWEs, PEMFCs, AWEs, and SOFCs) contributes to attain these goals since these devices enable a low-carbon energy economy for both transport and electricity, allowing the storage and use of renewable energy in form of hydrogen. For instance, FCH technologies offer a significant reduction in the emissions of particulate matter, CO_2, and NO_x. However, and despite significant progress in the last decade, economic issues still hamper the deployment of FCH technologies. Fostering the collaboration between research and industry is a priority, as they have to work together in order to promote strategies to make hydrogen technologies cost-competitive. In this respect, eco-design practices should be considered by manufacturers, *e.g.* increasing the share of recycled materials (with special attention to critical materials) and considering technical aspects such as material compatibility to increase the share of recyclable material and allow the reuse of components after EoL. In addition, to make FCH technologies ready for the market, lifetime is a key parameter to improve; in this respect, failure mechanisms of both stack and BoP components have to be prevented.

The aim of this work is to deliver a reference document paving the way for a more comprehensive guide to optimise the supply chain of FCH technologies. Therefore, the goal is to provide valuable information, since the design phase, to help manufacturers to optimise the life-cycle stages of FCH products in terms of cost-effectiveness and environmental concerns linked to emissions and waste management. An imperative action is the alignment with the relevant European directives [2], in particular the Eco-design Directive [3] and the WEEE Directive [4]. The WEEE Directive sets specific recovery and recycling targets for defined electronic waste categories, considering material limitations defined in the Directive on Restriction of the use of certain Hazardous Substances in electrical and electronic equipment (RoHS) [5] and in the Registration, Evaluation, Authorisation and Restriction of Chemicals (REACH) Regulation [6]. Additionally, the Eco-design Directive is a key reference to improve the environmental profile of products by addressing design parameters (*e.g.*, material selection and substitution) that promote benefits through the whole life cycle, *i.e.* from the design up to the EoL. These directives play a key role in the FCH sector, promoting not only sustainable development but also strategic business benefits (*e.g.*, enhanced company reputation and product image) for the actors involved in the FCH supply chain.

2. Structure of the work

The definition of comprehensive strategies for the EoL of FCH products is pursued in order to favour their deployment in accordance with the EU regulatory framework. First, the main actors involved in the supply chain of FCH products are identified and their roles are defined. Subsequently, new strategies applicable to the EoL of FCH products are classified on the basis of the role that the actors may play in short-, mid- and long-term scenarios of the FCH market deployment.

Strategies applicable at the design stage are also presented taking into account the Eco-design directive. As a key eco-design strategy, the potential reduction or replacement of critical materials is addressed for the different components of the devices addressed in the HyTechCycling project (*i.e.*, PEMFCs, PEMWEs, AWEs, and SOFCs).

3. Identification of actors

In the following subsections, the main actors involved in the supply chain of FCH products are presented. Relevant activities are also described.

3.1 Virgin material supplier

Virgin material suppliers carry out operations of extraction, synthesis or production of virgin raw materials such as polymers, plastics, metals from ore, catalysts, *etc*. In some cases this stakeholder uses significant amounts of recycled materials (*e.g.*, steel manufacturing uses a significant part of iron scraps in the blast furnace along with cast iron, carbon coke, and limestone). Nevertheless, the final product is conformed as a virgin material rather than a recycled one. As shown in Figure 1, suppliers sell virgin materials to FCH manufacturers or other industrial sectors (*e.g.,* construction, aeronautical, IT, *etc.*).

3.2 FCH components manufacturers

In the manufacturing of both stack and BoP components, producers buy raw materials (steel, polymers, steel, REEs, catalysts, *etc.*) from different suppliers. Raw materials can be virgin or come from recycling processes as long as their properties meet the specific technical requirements. These materials undergo several processes to obtain semi-finished products (or sub-components, *e.g.* electrodes or helix) that are eventually assembled in finished stack components (*e.g.*, MEA) or in the BoP component (*e.g.*, blower).

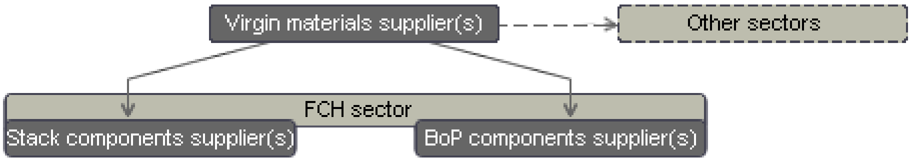

Figure 1. Suppliers of virgin material and components for the FCH sector

3.3 FCH system manufacturer

FCH system manufacturers (Figure 2) assemble stack components to obtain stack units, which are then integrated with the different BoP units to obtain the final FCH products. The supplier of BoP components (piping, cable, heat exchangers, blowers, *etc.*) is not necessarily the same of the stack components. Besides technical specifications, good environmental management practices should be provided, for instance, as documented eco-design practices, and suppliers and manufacturers should set sustainability goals at the company level to improve the performance of their products while favouring the circular economy concept. Typically, FCH manufacturers choose their component suppliers based on cost-effectiveness and design parameters. In this sense, sustainable practices applied (*e.g.*, the use of recycled material or the reuse of components) should also be provided to users.

3.4 User

The central actor of the product life-cycle is the FCH user. At the EoL, the final user could decide if the destination of the old FCH devices is a recovery centre (remanufacturer) or the waste manager for final disposal. Furthermore, the willingness to pay of users for FCH products is a crucial aspect for the FCH industry. For example, in the mobility sector, people in many countries are willing to pay for more powerful vehicles rather than for more efficient and less polluting ones [7]. In addition, when referring to FC vehicles, there are social concerns linked to safety that negatively affect the propensity of consumers to rely on hydrogen technologies, slowing down the transition to hydrogen. As long as this situation remains, the industry of mobility is not motivated to address investments towards these new technologies [8]. In general, along with technological advances and regulations, customer preference is a key driver determining techno-economic trends at the level of the whole industrial sector. Suitable tools for orienting consumer preferences are *e.g.* represented by sharing the information and knowledge transparently by the manufacturers, or by applying labelling and marketing strategies to promote sustainable approaches.

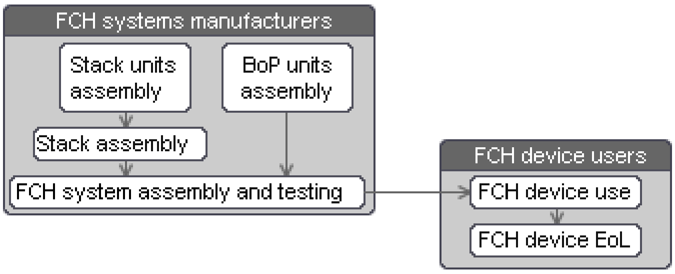

Figure 2. Main operations performed by FCH systems' manufacturers and final users

3.5 Recovery centre

The recovery centre (RC) is also a key actor at the EoL of products. RC actions stimulate the interaction and the exchange of secondary raw materials and components with manufacturers along the entire supply chain. Therefore, a fundamental role or RCs is fostering circular economy. In fact, the actions that the RC performs can lead to a significant reduction in costs, energy requirements, emissions, resources, and waste generated in the whole life cycle of FCH products.

As Figure 3 shows, once the RC receives the old devices and checks the general conditions and the main components to identify the mechanisms that determined the failure, the systems can be classified in recoverable or non-recoverable. Recoverable systems are partially (or, in some cases, completely) disassembled, and then the damaged components are repaired/substituted. The remanufactured system can be re-distributed to customers or to the distributor (which coincides with the FCH manufacturer) at a good price for both sides (win-win situation).

Regarding non-recoverable FCH systems, the complete non-destructive disassembly is a necessary practice to allow the direct reuse of as many components as possible (after simple operations of cleaning, if necessary). For non-directly reusable components, according to the European Waste

Framework Directive, waste management starts (Figure 4), prioritising preparation for reuse. In this sense, when economically feasible, operations of reconditioning, remanufacturing and repairing are performed at the level of components; then, the components are tested to re-enter the FCH market. The practice of reuse allows saving energy, costs, landfill space, and natural resources; non-reusable components and parts undergo mechanical and/or chemical treatments for the recovery of valuable materials. In part, these treatments can take place at the RC (according to the novel and/or existing technologies available [9,10]), which fosters closed-loop recycling (*i.e.*, use for applications in the FCH sector). Components and parts for which material recovery is not possible or is not economically convenient are collected by the waste manager.

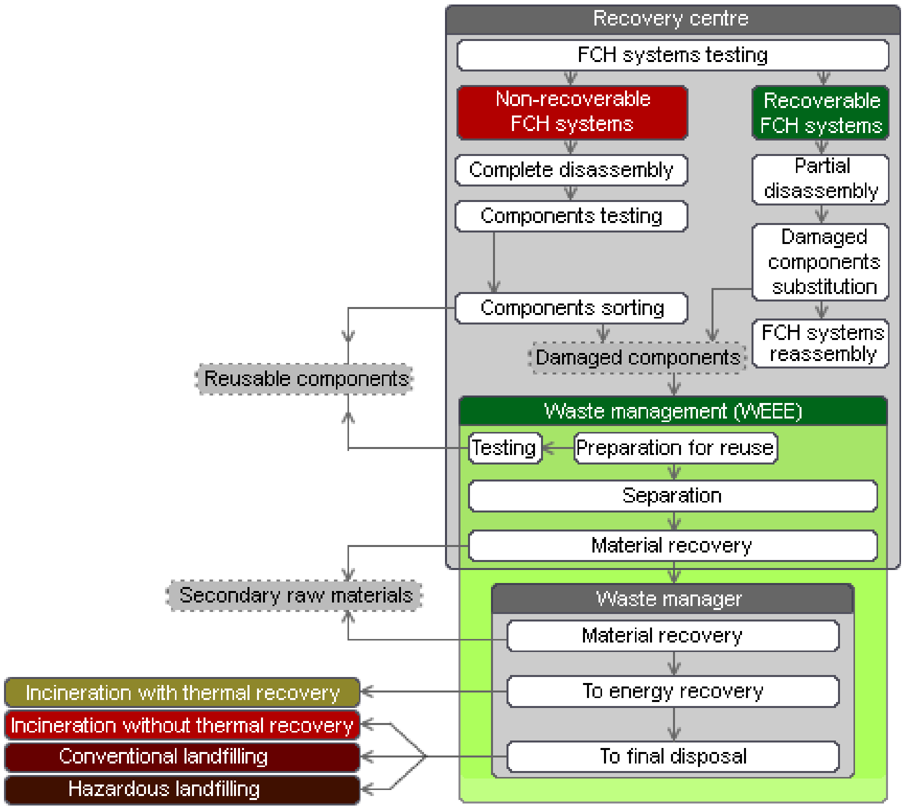

Figure 3. Main operations performed by the recovery centre and the waste manager

3.6 Waste manager

Finally, the waste manager, collects, sorts and delivers, in a safety way, all the remaining parts to the final destinations. From these parts, some materials can be additionally recovered through further mechanical and chemical treatments. The remaining parts for which material recovery is not economically or technically feasible are sent to final disposal, which can be incineration with energy recovery, incineration without energy recovery or, depending on the hazardousness of the material, dumping in conventional or hazardous landfills. According to the EU policies on waste management [11,12], and as defined in the Waste Framework Directive [13], priority is given to material recovery, followed by incineration with energy recovery and, when unavoidable, final disposal (Figure 4).

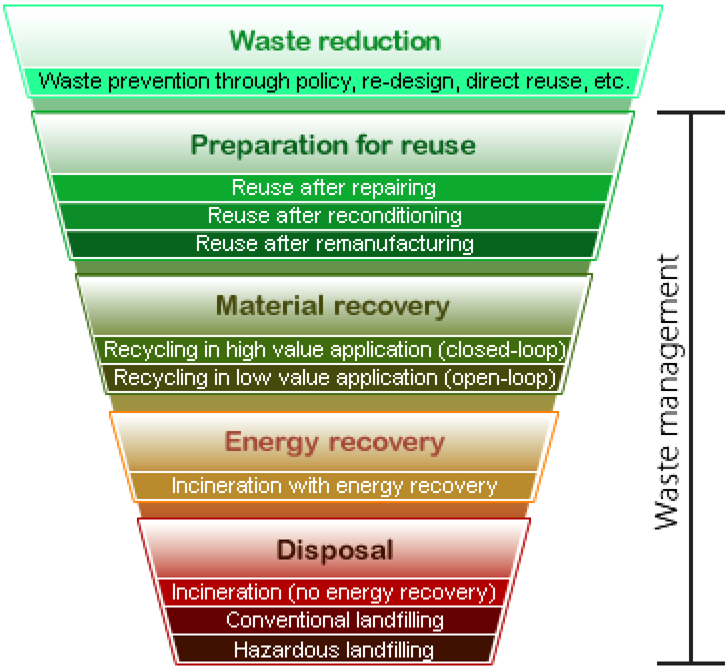

Figure 4. Waste management hierarchy

4. Classification of EoL strategies applicable to FCH products

4.1 Based on the role of the actors

Figure 5 shows the main flows and the interactions between the actors identified in the previous section. The core of the circular flows (green round-shaped arrows) is the recovery centre from which the circular flows branch off. It should be clarified that the figure does not emphasise the places where the actors are located (addressed in the following section), it rather presents the actors and the main actions they perform, with the aim of defining the most relevant criteria to identify FCH strategies.

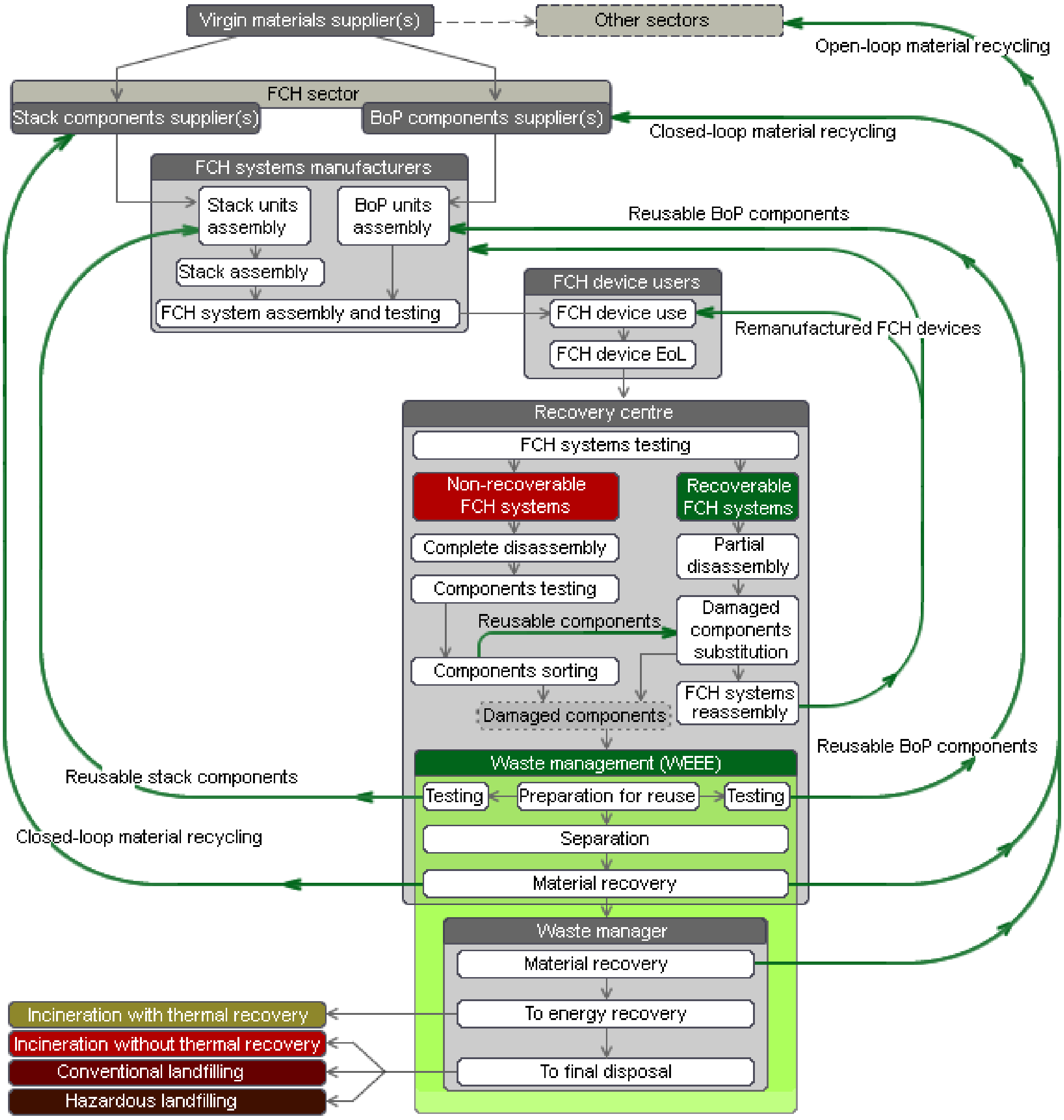

Figure 5. Main actors, actions and flows in the life cycle of FCH products

The operations performed by the FCH recovery centre allow the reduction of costs, waste to disposal and the related impacts. If specific recycling technologies are used [10], the reuse of stack components and the recovery of critical materials in a closed-loop scheme (*i.e.*, in the same sector or in equal high-value applications) would become possible. In addition, when components are directly reusable, the recovery center itself might reuse them as spare parts for reconditioning, remanufacturing or repairing entire FCH devices. Recovered devices could be directly sold to users or to the FCH supplier, which is supposed to be the original FCH manufacturer. In the overall picture in Figure 5, the smaller the circle of circular economy flows, the better can be considered the practice as higher amounts of waste, resources, costs and environmental concerns are avoided.

The role played by each of the actors is highly influenced by the development of the FCH market. In the short term, FCH applications are expected to remain limited to a niche market. It is assumed that the importance of the hydrogen is expected to grow in the future across many sectors, such as industry, energy, transport, and residential ones. Sector strategies based on the roles of the recovery centre, the waste manager and the FCH manufacturer are discussed below.

4.1.1 Short-term horizon

As anticipated, in the short-term, with a low grade of deployment of FCH technologies limited to few applications, users and waste managers are the main actors involved in the EoL stage. Figure 6 shows that the size of the RC (when included in the scenario) changes with the level of development of the FCH market and the concept applied (centralised/decentralised RC). The FCH manufacturer (which is supposed to correspond to the distributor of the devices) is involved in the trade of FCH devices and it might be involved in the extraordinary maintenance or, for large devices, in the ordinary maintenance. If the waste manager was specialised in FCH devices, reusable components would be recovered and re-sold to the manufacturer as spare parts. In this scenario (Figure 7), the transport of new devices from the manufacturer to the user should be paid by the user, as well as the transport to the waste manager. In the case of specialised waste managers, EoL transport could be paid by the waste manager because of the benefits derived from the recovery of components. Circular economy flows would be limited to the recovery of some types of plastics and metals in an open-loop scheme, while, in the case of a specialised waste manager, some components would be additionally recovered (Figure 5).

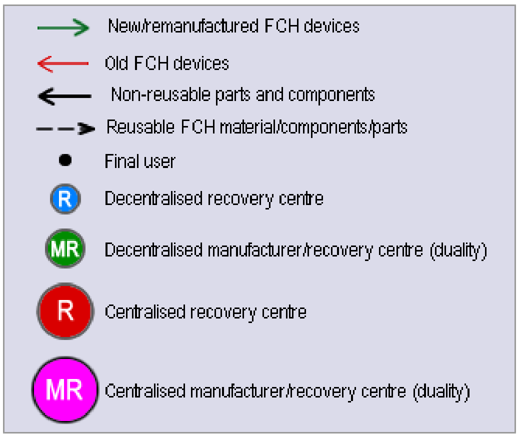

Figure 6. Legend for scenarios A-E

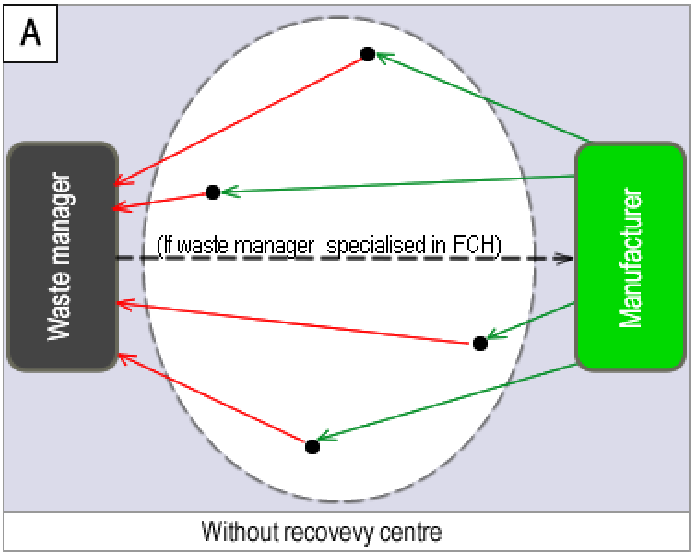

Figure 7. Scenario A (without FCH recovery centre)

4.1.2 Mid-term horizon

In the mid-term horizon, a growth of the market and a higher number of FCH users is expected. Despite this growth, as usual for novel energy technologies, in the mid term the market volume would only be partly settled (with a wider deployment expected in the long term). In this scenario, the role of specialised recovery centres in the EoL phase is fundamental to boost and support the deployment of hydrogen technologies. In the mid-term horizon, the EoL of FCH devices would represent a new business for companies and would be likely stimulated by a growing economic relevance of FCH products. In this scenario, focusing on the regional scale, two main alternatives are identied: (i) centralised recovery centre (Scenario B, Figure 8), and (ii) decentralised recovery centres (Scenario C, Figure 9). In both scenarios (B and C), the activity of the recovery centre would minimise the waste flow to the waste manager, reducing environmental concerns. This would promote the reuse of components and the recovery of valuable materials to re-enter the FCH market, reducing costs. Novel EoL strategies may start to be used together with existing ones for the recovery of PGM and other critical materials [9,10].

The transport of old devices to the recovery centre should be paid by the recovery centre itself, which would be balanced by the likely economic benefits. Alternatively, transport could be paid by the final user if stimulated *e.g.* by the reduction of taxes or economic inventives to buy a new (or remanufactured) device.

Finally, it should be noted that the centralised recovery centre should be able to manage all the old devices of a region, and therefore it would require larger structure with larger capacities. In contrast, in scenario C the single capacities of the decentralised recovery centres would be lower.

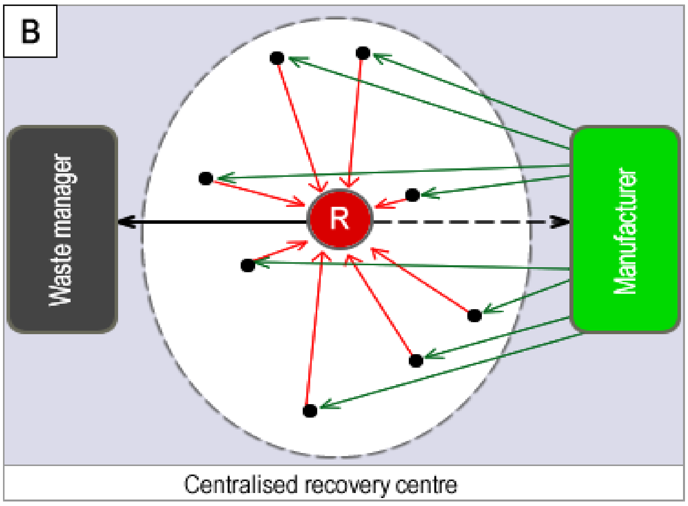

Figure 8. Scenario B (with centralised FCH recovery centre)

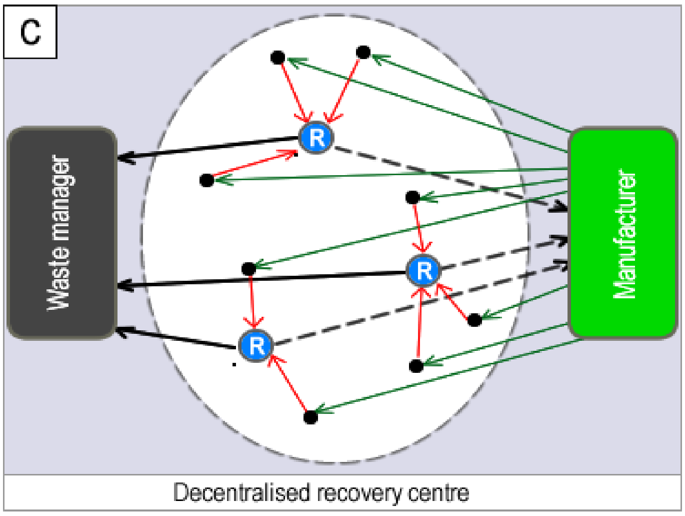

Figure 9. Scenario C (with decentralised FCH recovery centres)

4.1.3 Long-term horizon

In a more extended time horizon, under a decarbonised energy sector scenario, the hydrogen economy could be well established across regions and countries. This would imply the deployment of FCH products on different scales and for different applications, with a high number of final users. Within this context, it could be economically convenient for FCH manufacturers to perform different roles simultaneously (duality), *e.g.* recovery centre and distributor. The multiple role of a single actor would mean a higher control of the company over the products' life cycle, facilitating a complete optimisation of the supply chain. With a high market volume, a higher number of transport of devices should be expected, thus requiring logistic optimisation. In this sense, reverse logistics might be applied, *e.g.* for the transportation of old and new (or repaired/reconditioned/remanufactured) devices between the user and the manufacturer/recovery centre, reducing empty trips with economic benefits on the FCH business.

The two possible concepts, centralised (Figure 10) and decentralised (Figure 11), would also occur in this situation, but –due to the multiple role manufacturer/distributor/recovery centre (MR)– the flows of devices, components and materials change significantly.

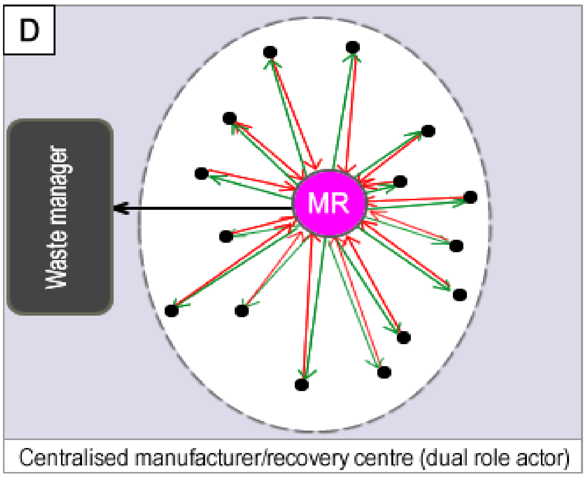

Figure 10. Scenario D (with centralised manufacturer/FCH recovery centre, dual role)

MRs in general have the possibility of performing FCH upgrading activities. Obsolete components would be directly reused in lower grade applications or as spare parts for repairing operations, promoting the fulfilment of the waste hierarchy. The reusable spare parts recovered through the activities of decentralised MRs (dashed lines in Figure 11) could be shared by the different centres when the availability of items collected is sufficiently high.

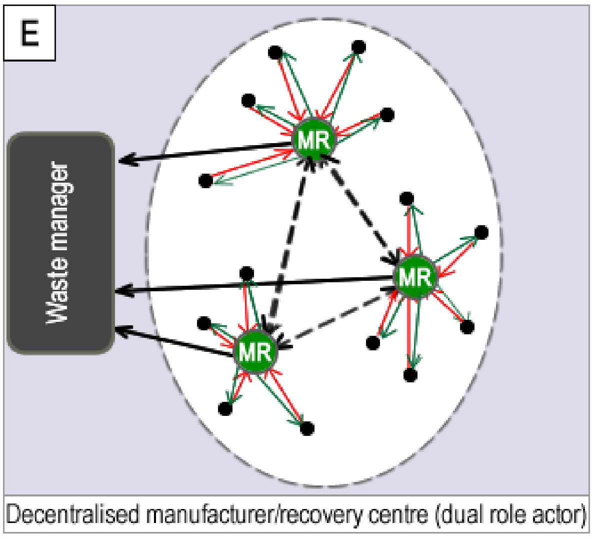

Figure 11. Scenario E (with decentralised manufacturer/recovery centre, dual role)

Extended producer responsibility (EPR) might represent a valuable tool to strengthen upgrading activities and mitigate the risk of loss of old devices, further promoting circular economy. In fact, EPR pushes manufacturers to recover FCH devices at their EoL, while manufacturers should encourage final users to give old FCH devices back. Therefore, in this scenario, EPR flanked by a new business model based on leasing instead of ownership, and with adequate marketing strategies to enhance product reputation, would represent a possible way to foster circular economy, waste prevention, economic feasibility and company image in the sector of FCH products.

4.2 Based on eco-design

The Eco-design Directive is a framework directive that helps EU to achieve the 20-20-20 targets. The Eco-design Directive applies to energy-related products with a sales volume above 200,000 units per year through the internal European market.

The Eco-design Directive specifies the main parameters that must be considered to improve the environmental aspects when designing a product. In particular, when selecting materials, it must be considered:

i) Reduction in weight and volume of the product. For example, when designing parts requiring high mechanical properties (stiffness, toughness, robustness, *etc.*), the achievement of the adequate properties by using better construction techniques (at the material supplier level) should be prioritised over oversizing the component (at the component manufacturing level). In FCH devices, it can be exemplified by end plates of SOFC or PEMFC stacks.

ii) Employment of materials issued from recycling activities. This practice allows both the improvement of the overall environmental performance and the increase in the regional demand for recycled materials. This recommendation is easily applicable to components made up of conventional metals and plastics (for which recycled raw materials are usually available) such as housing parts. One of the challenges for the FCH supply-chain actors is the use of secondary raw materials in the stack components, and in particular for catalysts, electrodes and membranes, which involve critical materials.

iii) Reduction in the consumption of energy, water and other resources throughout the life cycle, as well as the reduction in the level of emissions to air, soil, and water. To go in this direction, it is essential to look at the life-cycle stage in which the highest consumption of resources and energy takes place. In this sense, a detailed Life Cycle Assessment (LCA) would be convenient to quantitatively identify the critical stage(s) of the supply chain, including EoL.

iv) Incorporation of used components. This practice foresees the possibility of reusing units – directly or after a stage of preparation for reuse (*i.e.*, repairing, reconditioning or remanufacturing)– or components of the devices. Therefore, the core system, its sub-units and ancillary components must be assembled with procedures that allow a non-destructive and affordable disassembly in the EoL phase for possible repairing and upgrading activities. A higher rate of reusability can be reached through the accurate application of eco-design measures, prioritising the avoidance of technical solutions detrimental to the reuse and recycling of components and whole appliances. According to the state-of-the-art in FCH-oriented EoL technologies addressed in the HyTechCycling project [9,10], only BoP components (blowers, pumps, wiring, piping, *etc.*) could be reused. Regarding stacks, some components could be potentially reused only if novel EoL technologies were used [10]. This indicates that significant efforts towards technological advances are needed from the side of both manufacturers and recovery centres.

v) Design of durable parts for the extension of the lifetime as expressed through a minimum guaranteed lifetime, a minimum time for availability of spare parts, modularity, upgradeability, and reparability.

vi) Reduction of amounts of waste generated, with particular attention to hazardous waste. When avoiding the use of hazardous materials by manufacturers is not possible, designers must prioritise the use of these materials coming from recycling activities. In order to increase recyclability, the use of adhesives, paints and polishes has to be minimised, as well as the need for lubricants, fluids, and other consumables.

vii) Use of marking for parts in agreement with the ISO standards for plastics and rubbers, and when possible, identification of metal alloys. This makes the identification of materials easier, thereby enhancing their sorting and avoiding cross-contamination.

viii) Prioritisation of the use of compatible materials. FCH manufacturers should be willing to (re)design systems, devices and components looking for compatibility to allow full recovery of critical materials with new recycling processes. It is indispensable to guarantee, since the design phase, compatibility, so that one critical material can be recovered without affecting the likelihood of recovering others. In this respect, recycling compatibility charts are a useful tool to select suitable materials.

4.2.1 Reduction or replacement of critical materials and components

To promote the deployment of FCH technologies, the common key requirement is the cost-effectiveness of the devices. In this respect, high-volume production, technology innovation and supply chain development need to be pursued in addition to cost reduction. Regarding technology innovation, the material selection is a central action in the eco-design activity, and REACH and RoHS Directives have weighty repercussions on that. In particular, for FCH devices produced in Europe, where the scarcity of critical raw materials is important, it is necessary to explore new alternatives towards their reduction or replacement [14]. In this document, special attention is paid to PGM (mainly used in AWEs, PEMFCs and PEMWEs) and rare earth elements (REE; used mainly in SOFCs). The hazardousness of materials is also a key parameter taken into account. In this sense, Ni-based materials (mainly in SOFCs and AWEs) and materials for membranes (in PEMFCs and PEMWEs) are also relevant to this document.

4.2.1.1 Reduction or replacement of critical materials and components in SOFCs

Thanks to their high conversion efficiency (chemical energy of the fuel is converted into electricity and heat at high temperature), SOFCs find application mainly in the stationary market, for both small plants (few kW) and medium plants (many MW). Currently, a key challenge for SOFC manufacturers is to decrease the operating temperature (which causes severe degradation), while maintaining at least equivalent technical features. This enhancement would improve the life cycle of the devices from both environmental and economic perspectives. The challenge becomes harder when these improvements are addressed trying to use materials that are non-critical (or less critical) than the ones of the state-of-the-art.

Anode

The key requirements for SOFC anode are an excellent catalytic action for the oxidation of the fuel, an adequate porosity, and high stability and electrical conductivity. Nickel and nickel oxide shows suitable properties as the anode material, but its high thermal and chemical expansion coefficients make its use in combination with other materials necessary. In addition to this, nickel-based anodes show a low tolerance to sulphur, which narrows the range of suitable fuels. Furthermore, nickel oxide and metallic nickel are classified as a carcinogen (category 1) in the REACH Directive [6], therefore their use must be limited. Since existing technologies for the recovery of a high rate of nickel are available, policy-makers should encourage manufacturers of the electrochemical sector to prioritise the use of recovered nickel in high-grade applications over the use of virgin materials. Promising alternatives to the use of nickel in SOFCs are represented by full ceramic cells based on Ni-free anode such as $La_{0.20}Sr_{0.25}Ca_{0.45}TiO_3$, $Sr_{0.895}Y_{0.07}TiO_3$, $La_{0.2}Sr_{0.7}TiO_3$, $Sr_{0.94}Ti_{0.9}Nb_{0.1}O_3$, and $La_{0.3}Sr_{0.55}Ti_{0.9}Cr_{0.1}O_3$ [15,16]. In this novel SOFC concept, the anode material is based on titanium oxides, which, in SOFC devices, does not present particular criticalities in terms of costs, supply risk or toxicity [17]. In addition, these materials show a number of advantages such as a negligible chemical expansion (during redox cycling), a thermal expansion coefficient matching the one of zirconia electrolyte, a high sulfur tolerance, and reduced costs [16]. Titanium can be recovered at the EoL stage through conventional methods based on physical separation (size reduction, and magnetic separation), but, being combined with other elements, its recovery as anode material would require more complex processes, *e.g.* hydrometallurgical processes. Besides the aforementioned electrochemical requirements, mechanical features require anode materials with a lower sensitivity to the temperature and a thermal expansion coefficient matching the one of the adjacent components (interconnects and electrolyte layer) to avoid cationic interdiffusion at the interface. Currently, the most common SOFC anode material is a cement composite of YSZ powder as support of Ni particles as the catalyst. From the side of the cement material, though YSZ does not present concerns liked to hazardousness, its main criticality is linked to supply risk due to yttrium scarcity [18]. Therefore, actions for reducing its load in SOFC devices are necessary. In the EoL phase of SOFCs, YSZ can be recovered through existing hydrometallurgical treatments [9], but in the use phase, 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 as SOFC electrolyte). Alternatively, if hydrometallurgical recovery is not applied, the ceramic composite material can be recycled, after grinding and mechanical separation, still in an open-loop scheme, in the construction sector as a filler material or to remanufacture bricks [19]. The substitution of YSZ with non-critical (or less-critical) materials represents a very hard challenge, especially in conventional (high temperature, 800-1000 ºC) SOFCs. YSZ presents an excellent ionic conductivity at the operating conditions of SOFCs, preserving its good mechanical properties, gas impermeability, no electronic conduction, and stability in both oxidising and reducing atmospheres. Another important strength is that YSZ is highly compatible (chemically and mechanically) with adjoining components (anode and cathode) ensuring interfacial adherence in the operating range of temperature. For these reasons, YSZ is employed both in the anode compartment and as the electrolyte. A number of potential candidates can be thought for substituting YSZ in SOFCs as the electrolyte, but they present the same criticalities of YSZ since are based on rare earth dopants (scandium, samarium, ytterbium, gadolinium, *etc.*) [20,21]. Moreover, it is crucial to take into account that the selection of cathode and anode materials is extremely dependent on the type of electrolyte.

Cathode

Regarding the cathode side, the state-of-the-art material is LSM, but the replacement or substitution of La cations with Pr seems to be a suitable way to improve the features and the durability of the cathode. Due to promising oxygen transport properties and good catalytic activity, praseodymium-based perovskites structures are gaining interest. A potential solution for designing SOFCs with a reduced amount of these critical materials is represented by a lower operating temperature. In this respect, intermediate SOFCs (IT-SOFC) operate in the range of 600-800 ºC, while low-temperature SOFCs (LT-SOFC) operate between 300 ºC and 600 ºC. On the one hand, relevant advantages of LT-SOFCs and IT-SOFCs are the reduction of oxidative degradation and the possibility of using metallic interconnects instead of Ni-based alloys. Since the cost of interconnects accounts approximatively for more than one-third of the stack cost [22], this substitution has relevant benefits not only linked to the reduction of hazardous materials but also to the manufacturing costs. On the other hand, when operating at decreased operation temperatures, the main issues are represented by the polarisation of the cathode (made up of LSM in conventional SOFCs) with adverse consequences on electronic conductivity. For IT-SOFCs, the problem at the cathode side can be addressed by substitution of Mn by Fe or Co. It is important to underline that lanthanum-based cathode, being a heavy REE, suffers from the criticality of a higher supply risk than yttrium (light REE).

Electrolyte

For IT-SOFCs and LT-SOFCs, the problem of reduction in ion conductivity of the electrolyte can be addressed by designing electrolytic layers reduced in thickness and/or substituting the material. These measures will have the consequent benefit of critical material reduction [20]. For instance, metal-doped bismuth vanadium oxide (also known as BIMEVOX) electrolyte systems show good conductivity, but the instability in the reducing anodic condition is the main barrier for the application of this concept to SOFCs

[23]. This inconvenient is overcome in two-layer electrolyte concepts (made up of doped cerium oxide/YSZ layers and doped cerium oxide/stabilised bismuth layers), but this concept arises concerns on worse electrolyte performances and chemical and thermal stress. It is worth mentioning that further composite electrolytes based on ceria-carbonate are largely researched [23,24] due to their excellent ionic conductivity.

A potential improvement when designing LT-SOFCs is the possibility of recirculation of anode gas. It has been indicated that this measure increases the efficiency of the system by recycling waste heat and water [22]. Nevertheless, this method can be applied only at lower temperatures due to technical limitations of the blowers. By operating at a lower temperature, capital and operating costs reduction will also be achieved because of the lower requirements of BoP components (*e.g.*, heat exchangers, recyclable blowers, and insulation) and the possibility of manufacturing metal supported cells with thinner layers of electrolyte (<10 μm) [15]. Metal-supported cells are still at an early stage of development, but they are gaining research interest. Therefore, as also pinpointed in the SETIS Materials Roadmap [25], the reduction of the operating temperature and the application of anode gas recirculation in the system design might be an area of high interest. On the other hand, technical drawbacks such as deposition of coke have to be overcome to maintain good performances, being sulphur poisoning particularly important when lowering temperature. It is important to underline that, for improving the technical performance of SOFCs, it is crucial not only the engineering approaches to choose materials, but also how the components (cells, interconnect, current collector, and sealants) are brought together. In fact, it has to be taken into account that when the cells are stacked together, the high number of interfaces cell/current collector/interconnect arises concerns about sealing and contact. Undesired effects on material interaction and degradation may occur if particular attention is not paid to the electrical contact between interconnects and cells. In this regard, current concentration around the irregularities of surfaces lead to weaker contacts that could feed overheated spots, with undesired consequences regarding premature failure of components [26]. This is not a secondary issue, especially regarding the cathode compartment (characterised by oxidising atmosphere), for which different approaches are possible. A common approach to deal with this issue is, for instance, the application during the assembly of a wet paste made up of cathode material; the viscosity of this paste will level the irregularities improving the reliability of the electric contacts. However, in the operative phase, the high temperatures would rise problems linked to delamination and cracking of the conductive layer. Moreover, it has to be carefully dosed since any excess could threaten the regular flows of gases or create short-circuits.

Overall, although a number of solutions involving alternative non-critical materials for SOFCs with good technical features are identified, further research is needed to better understand the most suitable materials for the deployment of these devices. Regarding economic feasibility, it has been demonstrated that Ni-based stack components are cheaper than lanthanum-based ones, but the quantity needed for Ni-based stacks is higher [22], and therefore more concerns regarding hazardousness arise. The complexity of the whole picture increases dramatically when more types of materials are considered as potential candidates, and when socio-environmental aspects are taken into account for the identification of the best eco-design solutions. Within this context, multi-criteria decision analyses could be suitable tools for a more reliable identification of electrolyte/electrode material selection in SOFC stacks encompassing economic, environmental, social and technical parameters following a life-cycle perspective.

4.2.1.2 Reduction or replacement of critical materials and components in PEMFCs

Thanks to their high power density, low operating temperature and low weight, PEMFCs can find application in mobility, in the portable market, and in stationary production. In PEMFCs, the use of critical materials is mainly linked to the manufacturing of electrodes. Among noble metals, the most active species are PGM [27]. In particular, Pt represents the state-of-the-art as the electrocatalyst in low-temperature PEMFCs for both the anode and the cathode. Due to the slower kinetics of the oxygen reduction reaction (ORR) of the cathode, the load of platinum is often higher at the cathode than at the anode. Depending on the application, the catalyst loading can vary from about 0.13 mg PGM/cm^2 of active area (0.16 mg PGM/kW with a power density of 0.8 W/cm^2) in transport application [28], up to *ca.* 0.4 mg PGM/cm^2 (1.5 mg MPG/kW with a power density of 0.27 W/cm^2) for power production [29]. Due to low operating temperature (about 80 °C) and the acid environments for the presence of H^+ ions, catalysts with high activities (typically Pt) are necessary for good performance of fuel cell stacks.

Figure 12, based on DoE information [29], shows the cost breakdown per net kW for 100 kW and 250 kW PEMFC stacks for stationary application at different production volume, with focus on material costs (BoP components are not considered). For each scenario, the contribution of MEA results significant, but, depending on the production volume, the single MEA component has different economic influence. On the one hand, at current low volume production, the costs of the membrane (Nafion DE-521 0.2 mm thick, PTFE reinforced) and GDLs (assumed to be purchased in roll form, made up of 0.2 mm thick carbon paper dip-coated with PTFE for water management) account for 30% each. Despite the fact that the main barrier is the need to use platinum catalysts for the reactions, which are both expensive and scarce, the cost of the catalyst (0.267 mg Pt/cm^2 and 0.133 mg Pt/cm^2 for cathode and anode, respectively, on carbon black support) accounts for less than 20% for both 100 kW and 250 kW stacks. On the other hand, in large volume production, the contribution of GDLs and membranes becomes almost negligible, whereas catalyst material accounts for about 45% of the total stack costs. This suggests that, depending on the scale of production, the strategy for lowering the stack cost should be different. Regarding GDLs, the production cost can be reduced significantly by re-engineering the manufacturing process, eliminating lengthy batch processing and multiple coating passes without a reduction in performance [30].

A way to improve the economic feasibility of the PEM stack through the optimisation of MEA manufacturing parameters (by hot-pressing) was proposed by Okur *et al.* [31]. The analysis consists in finding the optimum point (pressure, temperature, and pressing time) to achieve the maximum power density. The test was carried on symmetric MEA composed of Nafion 212 membrane sprayed with 40% Pt/C catalyst ink with a Pt load of 0.7 mg/cm². At the end of the experiment, the authors found two different optimal points, the first at 97 °C, 66 kg/cm² and 3.56, for which a power density of 862 mW/cm² was found; the second at 87.35 °C, 47.71 kg/cm² and 1.15 min with a power density of 768 mW/cm². While the former maximises the current density, with techno-economic benefits during the use phase, the latter minimises the manufacturing costs, promoting low-cost production of MEAs.

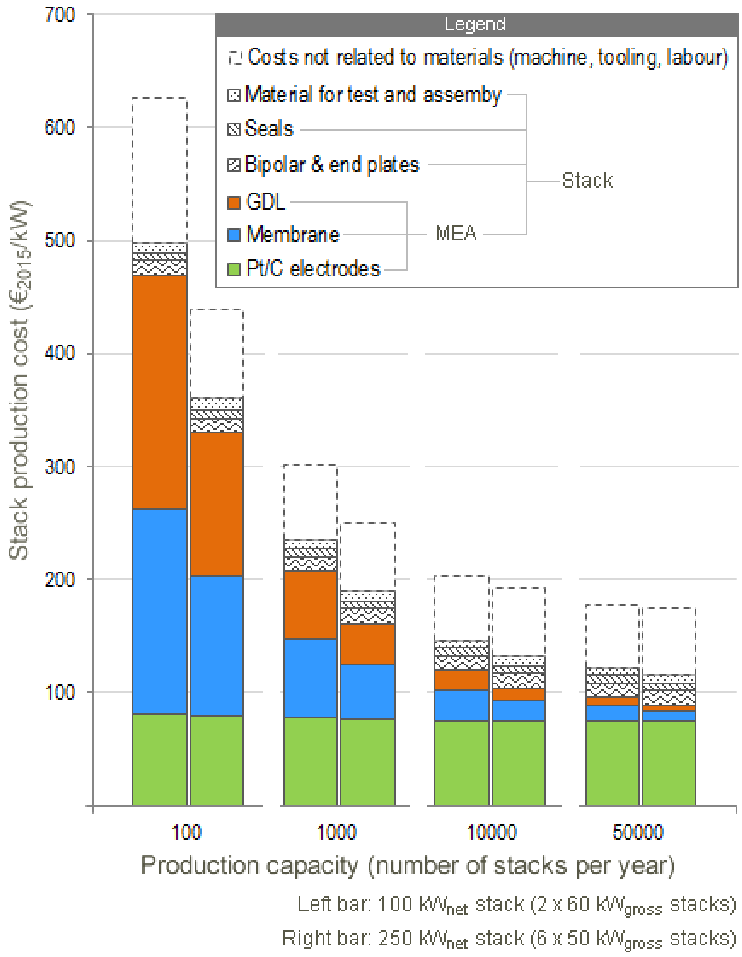

Figure 12. PEMFC stack costs breakdown (based on [29])

At the level of stack component manufacturer, technological advances can be made in order to reduce the amount of scrap. For instance, Muruganantham *et al.* [32] presented a study based on simulation models about the possibility of reducing significantly the loss of electrode material by enhancing the MEA manufacturing process with the inclusion of a scrap dismantling unit in the production chain.

PEM catalysts

The durability and the efficiency of PGM as the catalysts can be enhanced by substituting the support materials. The conventional material for catalyst support is carbon black. On the one hand, this material offers a high surface area for the catalyst, which allows a higher catalytic activity. On the other hand, carbon black corrodes, and the catalyst may detach from its support aggregating in a larger particle, with negative consequences on the active surface, and then on the cell's performance. In this regard, numerous conductive materials (*e.g.*, conducting polymers, ordered mesoporous carbon, graphitic nanofibre, carbon nanotubes, and graphene) are seen as promising alternatives. Nonetheless, due to higher costs and low durability of these novel materials, more research efforts regarding their manufacturing are needed for a cost-effective PEMFC production on a large scale [33]. The use of an appropriate support can reduce the quantity by enhancing the dispersion of Pt on the electrodes. Another possible way to achieve the reduction of quantity of Pt is to adopt the so-called core-shell concept, involving a structure of the catalyst with a core of non-Pt metal (or Pt alloy with lower cost than pure Pt) surrounded by a Pt shell (Figure 13), which can be synthetised via electrodeposition, leaching or redox exchange [34]. The performance using Pt alloys containing iron or vanadium shows a high activity for oxygen reduction at the cathode, while Cu-cores show high performance at the anode [35]. Another method proposed by Fofana *et al.* [36] shows promising cathodic performance of MEA. The method consists in lowering the Pt loading to 0.05 mg/cm^2 by a multilayer catalyst sputtering, adopting a GDL containing a microporous layer. Another MEA manufacturing technique allowing a sensible decrease of Pt load is based on pulsed-laser-deposition (PLD) on the GDLs or directly on the Nafion membrane. The study of Mróz *et al.* [37] shows that high performance of PEMFC stacks are possible (power density up to about 190 mW/cm^2 with further potential room for improvement) by adopting PLD for ultra-low Pt loading (0.18-7.44 µg/cm^2) when the catalyst is deposited on the GDL through this technique.

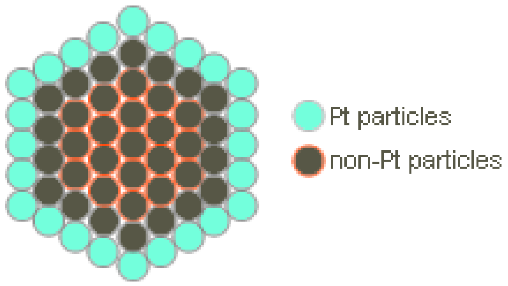

Figure 13. Core-shell catalyst structure

The recent experimental results obtained by Ye *et al.* [34] on a novel CCM design concept show the increase in cell performance (about 11% more in power density) reducing the platinum consumption to 0.29 mg/cm^2 (0.2 mg/cm^2 at the anode layer and 0.09 mg/cm^2 at the cathode). The novel CCM concept proposed (Figure 14) consists of a double cathode catalyst layer with a different density of porosity and platinum load, prepared via filtration method on the Nafion membrane.

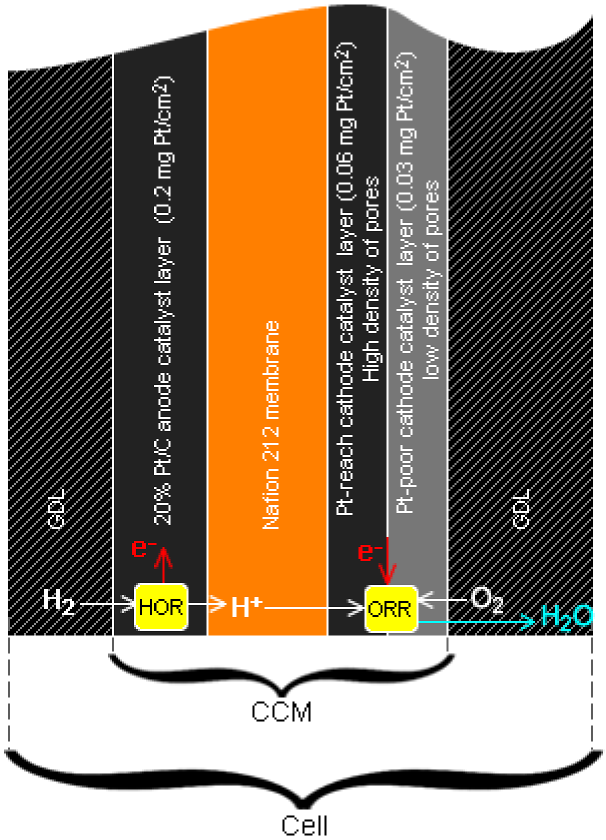

Figure 14. Dual-layer cathode catalyst layer in PEMFCs (based on [34])

Regarding substitution of Pt, Nabae *et al.* [38] reported the development of a Pt-free PEM cathode applying a catalyst prepared through multi-step pyrolysis of polymers containing nitrogen together with phenolic resins as the polymer precursors. The Pt-free catalyst shows appropriate features in terms of activity, stability and durability, proving to be a candidate to substitute conventional catalysts in PEM stacks.

Several types of Pt-free catalyst for PEM stacks are gaining interest. In particular, due to their high activity, Ni-, Fe- and Co-based electrocatalysts are potential candidates to substitute Pt on both electrodes. Regarding the cathode side, where the ORR takes place, MeN_xC_y catalysts are proposed in the literature as promising alternative catalysts [34].

PEM membrane

New alternative materials for substituting Nafion (which represents the state-of-the-art in PEM stacks) are highly desired, lowering the cost of the membrane while reducing concerns linked to Nafion disposal or incineration. The membrane material is essential as it requires being thermally and chemically stable and compatible with the adjacent part (GDL and electrodes). The 3M Company has undertaken a project to develop a new anion (instead of a proton) exchange membrane technology with potential applications in fuel cells and electrolysers [39]. The membrane would operate in an alkaline environment, allowing the use of lower-cost catalysts. PEM stacks work commonly in an acid environment at a temperature about 80°C and the key requirement of the membrane is to be able to conduct protons at the working conditions. Since working at a higher temperature would make substituting Pt with other catalysts possible, materials that allow working at higher temperature are particularly interesting for both research and industrial purposes. Several types of membranes for operating temperature above 100 °C have been developed, and can be classified into: (i) basic polymer doped with phosphoric acid, (ii) heterocyclic atmospheric proton conductors, and (iii) phosphonic acid proton conductors [35].

For the first category, several blends have been proposed, showing good features in terms of proton conductivity and long-term stability. The main disadvantages are represented by their mechanical stability and deterioration. Moreover, H_3PO_4 molecules come out of the membrane when operating at a temperature below 100°C.

Heterocyclic amphoteric proton-conducting systems are based on imidazole ($C_3N_2H_4$), which is an organic compound that liquefies in the range 91-256 °C. In this range, thanks to its high self-diffusion coefficient, it may be used as a proton solvent/conductor. However, its application on commercial scale is still limited by technical disadvantages such as the high over-potential for the ORR and low thermo-oxidative stability.

Similarly to phosphoric acid, ionomers containing phosphonic acid are able to conduct protons at a temperature over 100 °C without the presence of water. Many membranes based on this material have been presented at the research level, and the results show promising technical features, overcoming the disadvantages of basic polymer doped with phosphoric acid regarding thermal and mechanical stability. Nevertheless, improvements are needed to reach their commercialisation. In this sense, scientific groups have reached interesting results, achieving higher proton conductivity and power density with a membrane based on silica particles for composite membrane, prepared by inserting polystyrenesulfonic acid-grafted silica particles into an inert polymer matrix of poly(vinylidene fluoride-co-hexafluoropropylene) [39].

4.2.1.3 Reduction or replacement of critical materials and components in PEMWEs

Since the MEA components of PEMWEs and PEMFCs are based on the same compounds, and as the technical requirements are similar in these stacks, the considerations for PEMWEs related to the substitution of critical materials correspond to those for PEMFCs. A distinction can be made for bipolar plates, which in PEMWE are based on titanium. As Figure 9 shows, capital costs for a PEMWE system are estimated to be close to 2100 €$_{2014}$/kW (twice the costs of an AWE system) [40,41], and bipolar plates dominate the cost of a standard stack configuration (more than 50% of the cost of PEMWE stack) [40–42]. Since the production of titanium requires expensive and low efficient processes [43], the substitution of titanium with *e.g.* zirconium can provide a substantial benefit in economic terms. In this sense, many research activities and projects focus on the possibility of replacing titanium in PEMWEs with less expensive materials, maintaining the performance targets. Promising candidates have been found in refractory metals [44–46]. The cost of the electrolyte added to the cost of the electrodes can limit the development of PEMWEs; critical materials such as catalysts and membrane (Nafion) account for 8 and 5% of the total stack costs, respectively. The ELECTROHYPEM project found alternative materials for catalyst and membrane. For instance, a reduction in the electrolyte costs of 47% has been achieved with the new membrane made of reinforced Aquivion. The project addressed also the reduction of catalyst load (Ir and Ru) at the anode (from the typical 3 mg/cm^2 to 0.3 mg/cm^2). Due to the reduction of catalysts, it is possible to operate at higher current density (moving from 1 A/cm^2 to 3 A/cm^2) with benefits on the techno-economic performance of the entire stack. At the cathode, Pd and Pt were considered, with a reduction of catalyst load from 1 mg/cm^2 to 0.1 mg/cm^2, with relevant benefits in terms of costs and reduction of critical materials. The novel PEMWE system can obtain a reduction of costs about 11%, but the main challenge for the reduction of costs remains the substitution/reduction of bipolar plates' materials with new PEMWE concepts.

4.2.1.4 Reduction or replacement of critical materials and components in AWEs

AWE is a more mature technology than PEMWE; it exhibits more durable systems and lower capital costs thanks to the avoidance of noble metals in the stack. The criticalities of AWE materials are mainly linked to the harsh working conditions related to KOH as the electrolyte, and the hazardousness of Ni as the main material for electrodes. Recent studies [40–42] show that, at the level of the overall system, the AWE costs are equally distributed between stack and BoP components (Figure 15), and a significant cost reduction for AWEs could come from the possibility of reusing BoP components. The stack of AWEs is generally composed of Ni (for the electrodes), copper, and steel. For these materials, the recovery technologies are well-known. In the oldest AWE devices, it could be possible to find asbestos as membrane material. Since this mineral is harmful and carcinogen, it has been banned in the UE in 2005 [47]. However, several old devices are still working with asbestos membranes, and, because of the use of chrysotile in the diaphragm production, their authorisation to work can be extended until 2025. In developing countries, where the asbestos has not yet been banned, asbestos membranes can be thermally processed to produce harmless materials such as silicate glass or ceramic bricks. New electrolysers employing non-asbestos membranes make use of materials that are normally confidential, but, as for asbestos membrane, the main challenge for the EoL of AWEs is the possibility of recovering gaskets and the membrane. The technical requirements of the AWE diaphragm are high hydrophilic properties, chemical and mechanical stability in the alkaline environment, and economic feasibility. One of the most popular materials used for this purpose, Zifron, has been developed by VITO Research and commercialised by Agfa Group [48]. Zifron is based on 85% zirconium oxide, which provides the hydrophilic requirements, and 15% polysulphone network, for mechanical strength.

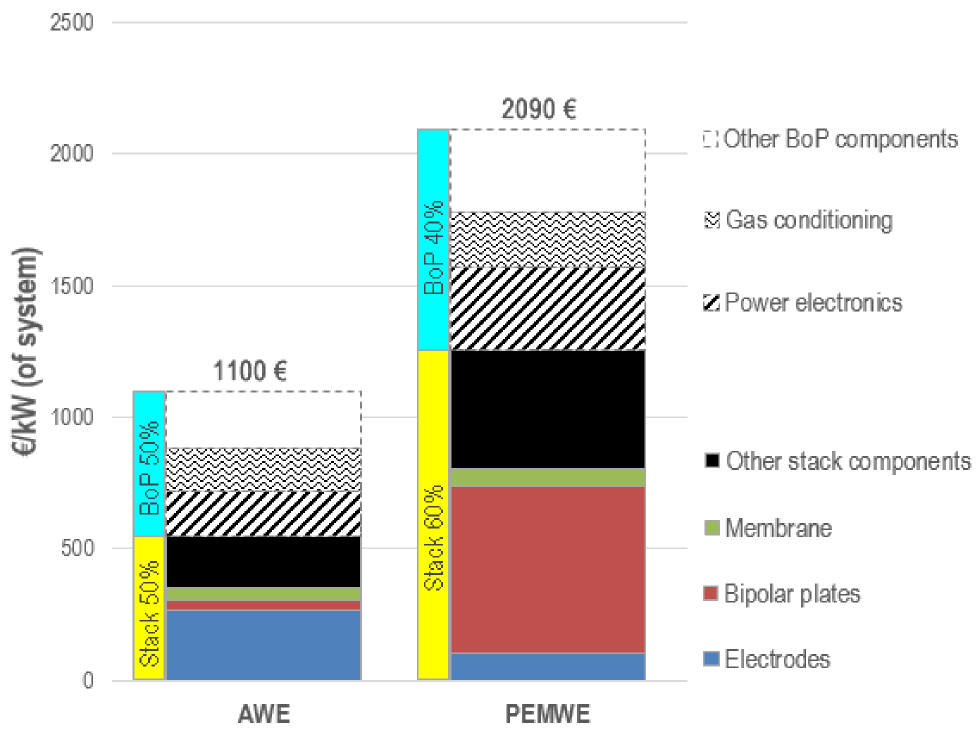

Figure 15. AWE and PEMWE systems cost breakdown (based on [41])

4.2.2 BoP components

For all the devices under study, and especially for AWEs, at the system level, the breakdown of costs shows that the BoP has a significant contribution. The number and type of BoP components, as well as the materials needed, are strongly dependent on the technologies and the technical parameters of the stack (temperature, gases humidity, current density, *etc.*). The key strategy to address BoP units is to design FCH systems allowing the reuse of as many BoP components (cable, piping, and other components such as blower and compressor, heat exchangers, *etc.*) as possible. These components should be easily accessible for many reasons such as their substitution in case of failure or system upgrading, their recovery at the EoL phase and their preparation for reuse. In this sense, FCH components located in a visible area are preferred over covered or hidden areas. Regarding stacks components, accessibility in direct axial direction needs fewer efforts than accessibility in radial direction. The number and the type of joints also affects the disassembly (snap fits are preferred over fasteners), or even a higher number of type of joints increase the difficulty in non-destructive disassembly since a higher number of tools would be required.

5. Conclusions

This study has identified potential room for improvement regarding FCH products along their life cycle, achievable through an optimisation of the EoL stage of these devices. The strategies identified according to the role played by the stakeholders take into account the localisation of recovery centres and manufacturers, size, and volume of the market in the short, mid and long term. For instance, in the short-term horizon, with a weak FCH market, conventional WEEE managers could enhance their level of specialisation in FCH to recover further valuable components and some materials in close-loop recycling through existing EoL technologies. In the mid-term, the deployment of specialised recovery centres that start employing novel EoL strategies represents a suitable strategy. In the long-term horizon, assuming a high level of deployment of the FCH technologies for different applications (residential, portable, *etc.*), logistic optimisation would be required to consolidate the FCH market.

Strategies potentially applicable in the short term also relate to eco-design practices. In this regard, after an extensive literature survey, strategies for the reduction or replacement of critical materials have been identified for all the devices considered in the HyTechCycling project. Regarding eco-design, the selection of the materials for manufacturing FCH products should follow criteria that minimise criticalities through the entire product's life cycle. In this sense, the most suitable materials should be identified based not only on cost minimisation but also on the energy demand for their extraction/production/synthesis, emissions, and use of hazardous reactants. It has been found that the level of development of the FCH market strongly affects manufacturers when deciding the material that should be reduced to improve the system's economic feasibility. According to the eco-design principles, manufacturers should prioritise recycled and/or recyclable materials (preferably in a closed-loop scheme), whereas the use of compounds dangerous for humans and ecosystems must be limited along the life cycle. For instance, regarding Ni used in SOFCs and AWEs, despite the advantages of high availability and relatively low price, its hazardousness (carcinogen) makes the use of alternative materials (maintaining appropriate technical features) necessary. In order to allow the reuse of FCH components as well as the use of recycled critical materials, it is necessary –during the design of FCH systems– to adopt measures that facilitate the non-destructive disassembly of the systems. Accessibility to every component has to be taken into account since it allows a less expensive and more effective EoL process, stimulating the reuse of components directly in the FCH products' supply chain.

Overall, although a number of solutions involving alternative non-critical materials and EoL technologies have been identified, further research is still needed to elucidate the most suitable strategies to be applied under economic, environmental and social aspects.

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