Sustainable SoluTions FOR
recycling of end-of-life Hydrogen
technologies

TRAINING MODULE 1

Chapter_1

Introduction to the FCs: main components and valuable materials in PEM and SOFC

Document Details

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Input from	Literature and Consortium

Document Details

x PU - Public
CO - Confidential, only for members of the consortium (including the EC)

Clean Hydrogen Partnership

Co-funded by the European Union

This project has received funding from the Fuel Cells and Hydrogen 2 Joint Undertaking (now Clean Hydrogen Partnership) under Grant Agreement No 101007216. This Joint Undertaking receives support from the European Union's Horizon 2020 Research and Innovation program, Hydrogen Europe and Hydrogen Europe Research.

Abbreviations

CEA	CEA research centre
CCM	Catalyst Coated Membrane
CHP	Combined Heat and Power
CL	Catalyst Layer
Co	Cobalt
FC	Fuel cell
GDL	Gas Diffusion Layer
GFCs	Grooved gas flow channels
HOR	Hydrogen Oxydation Reaction
HRD	Hensel Recycling
IDO	IDO-Lab
La	Lanthanum
LSCF	strontium-doped lanthanum cobaltite ferrite
LSM	lanthanum manganite
MEA	Membrane electrode assemblies
MPL	Micro Porous Layer
Ni	Nickel
Ni-GDC	nickel-gadolinium doped ceria
ORR	Oxygen Reduction Reaction
PEM	Proton Exchange Membrane
PFSA	perfluorosulfonic acid
POLITO	Politecnico di Torino
Pt	Platinum
PTFE	Polytetrafluoroethylene
RES	Renewable Energy Sources
SO	Solid Oxide
YSZ	Yttria-stabilised zirconia

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Contents

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1 Executive Summary

Within project BEST4Hy, a training kit is to be developed. This will be subdivided into 4 Modules with this content:

- Module 1: How to dismantle a fuel cell
- Module 2: Recovery technologies
- Module 3: Technical results and economical aspects
- Module 4: Measures towards to take up

Module 1 is dedicated to the dismantling technologies of fuel cell stacks and provides information on how to perform and optimize the existing disassembling technologies and how to perform novel dismantling technologies, considering both PEM and SOFC fuel cells.

Module 1 is organized as follow:

- ➢ Chapter 1: Introduction to the FCs: main components and valuable materials in PEM and SOFC;
- ➢ Chapter 2: Description and manual to perform the existing disassembling technologies (PEM);
- ➢ Chapter 3: Description and manual to perform the novel dismantling processes developed within the project (PEM).

Video tutorials on how to perform the disassembling and dismantling processes above described are also available.

For more information, users can consult the associated deliverables describing the activities performed in BEST4Hy:

- D1.1: Lab scale optimization results on the 3 PEMFC recycling technologies report
- D1.2 Technical report on adaptation of existing technology (hydrometallurgy process) for PEMFC material recovery: results and design
- D2.3: Report on the evaluation of MEA including recycled materials in small single cell of PEMFC
- D3.3: Pilot-scale plant (TRL5) based on two integrated existing recycling technologies for SOFCs_PU
- D4.3: Technical report on developed recovery technologies for LSC cathode materials_PU.

The present document refers to Chapter 1: Introduction to the FCs: main components and valuable materials in PEM and SOFC.

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2 Introduction

BEST4Hy (SustainaBlE SoluTions FOR recycling of end-of-life Hydrogen technologies), funded by the Fuel Cells and Hydrogen 2 Joint Undertaking (now Clean Hydrogen Partnership) within the European Unions' Horizon 2020 research programme, aims to find sustainable solutions for the recycling of end-of-life hydrogen fuel cells and their components.

BEST4Hy focuses on the development and validation of existing and novel recovery and recycling processes for two key fuel cell and hydrogen products: proton exchange membrane fuel cells (PEMFC) and solid oxide fuel cells (SOFC). A representation of the approach is provided in Figure 1.

The disassembly/dismantling phase has been explored to ensure maximization of material recovery.

Re-manufacturing of new cells / stacks includes:

- min 30% of recycled critical raw materials (Ni, YSZ and LSC) in SOFC cells manufacturing.
- min 95% of Platinum[1] in the manufacturing of PEMFC stacks.

Materials are evaluated for quality and performance in remanufactured PEMFCs & stacks and SOFC, so to deliver a concrete validation of the circularity potential within the hydrogen device industry.

In addition, BEST4Hy aims to maximize the recovery of hazardous materials such as nickel, platinum, yttrium stabilized zirconia, lanthanum and cobalt by establishing the degree of reusability in fuel cells (closed loop recycling) but also by exploring redirection to other markets (open loop recycling).

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[1] Ionomer could not be included in the remanufacturing of fuel cells due to difficulties in characterising the ionomer recovered

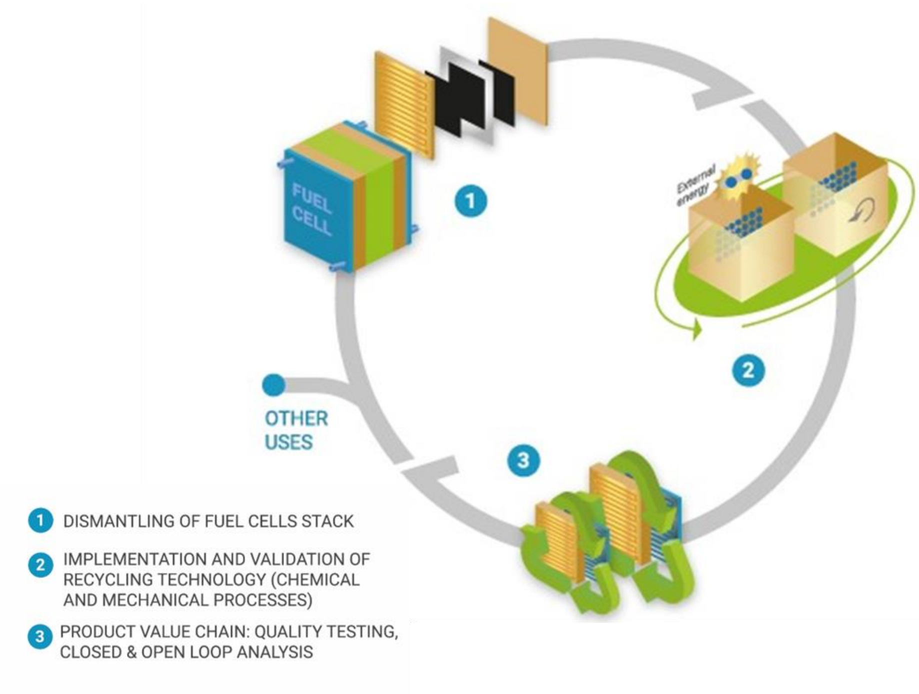

Figure 1 BEST4Hy approach

The different phases have some specific focus:

1. Disassembly & dismantling of fuel cells stacks: Maximization of material recovery, including improvement of dismantling procedures to reduce stack material losses of 5-10% (especially Pt) and recovery of a higher variety of materials; preliminary assessment of potential mechanization/automation to increase speed of operation.
2. Implementation and validation of recycling technology (chemical and mechanical processes): Recovery of critical raw and precious materials such as
   - Pt and ionomer from PEMFCs (HRD and CEA),
   - Ni and YSZ from SOFCs (PoliTO).

   All materials recovered will be fully characterized to maximize closed loop recycling.
3. Product value chain: Quality testing closed and open loop analysis. The materials obtained from the previous steps can be used for recycling into fuel cell products (closed loop) especially into MEA or to the secondary raw materials market (open loop).

3 Introduction to the fuel cells [1]

A **fuel cell is an electrochemical cell** that converts the chemical energy of a fuel (often hydrogen) and an oxidizing agent (often oxygen) into electricity through a pair of redox reactions. Fuel cells are different from most batteries in requiring a continuous source of fuel and oxygen (usually from air) to sustain the chemical reaction, whereas in a battery the chemical energy usually comes from substances that are already present in the battery. Fuel cells can produce electricity continuously for as long as fuel and oxygen/air are supplied.

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There are many types of fuel cells, but they all consist of an anode, a cathode, and an electrolyte that allows ions, often positively charged hydrogen ions (protons), to move between the two sides of the fuel cell. At the anode, a catalyst causes the fuel to undergo oxidation reactions that generate ions (often positively charged hydrogen ions) and electrons. The ions move from the anode to the cathode through the electrolyte. At the same time, electrons flow from the anode to the cathode through an external circuit, producing direct current electricity. At the cathode, another catalyst causes ions, electrons, and oxygen to react, forming water, heat and possibly other products.

Individual fuel cells produce relatively small electrical potentials, about 1 volt in open circuit down to 0.6 volts with load applied, so cells are "stacked", or placed in series, to create sufficient voltage to meet an application's requirements. In addition to electricity, fuel cells produce water vapor, heat and, depending on the fuel source, very small amounts of nitrogen dioxide and other emissions.

Fuel cells are classified by the type of electrolyte they use and by the difference in startup time ranging from 1 second for proton-exchange membrane fuel cells (PEM fuel cells, or PEMFCs) to 10 minutes for solid oxide fuel cells (SOFC).

Table 1 Comparison between PEMFC and SOFC

	PEMFC	SOFC
Applications	• Transport as a major application, stationary and portable power generator and electronic devices	• High-temperature (solid oxide) fuel cells more applicable for systems that run for extended periods of time without frequent start and stop cycles
Advantages	• Hight efficiency • Fuel flexibility • Smaller and less expensive than materials required for SOFC	• Can use CO+ Hydrogen as fuel without ar issues • Current technology can produce cheaper and lighter insulators (ceramics) • Better thermal management

PEMFCs are low temperature fuel cells that use a solid polymer in the form of a solid phase proton conducting membrane as an electrolyte which eliminates the need to contain corrosive liquids. PEMFCs are characterized by low temperature operation, high power density, fast start up, system robustness, flexibility of fuel type (with reformer) and reduced sealing, corrosion, shielding or leaking concerns.

PEMFCs are used in a wide range of portable and stationary power applications which include transport as a major application, stationary and portable power generator and electronic devices.

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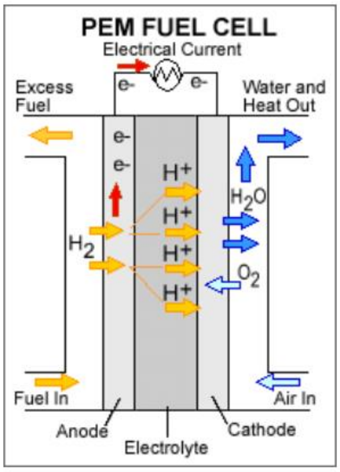

Figure 2 Polymer electrolyte fuel cell (PEMFC) schematic

Solid Oxide Fuel Cells (SOFCs) use a solid oxide material as the electrolyte and typically work at high-temperature. They are suited for systems that run for extended periods of time without frequent start and stop cycles. These systems also have benefits for CHP generation, and they offer simplified operation on fossil and renewable fuels. The high-temperature systems can also be utilized in tri-generation mode to produce electrical power, heat, and hydrogen.

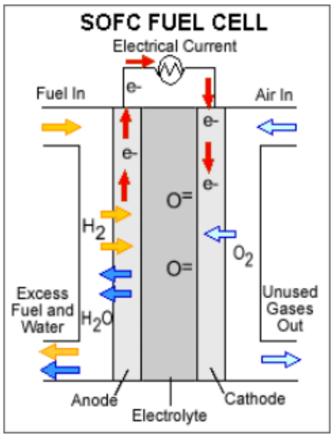

Figure 3 Solid Oxide fuel cell (SOFC) schematic

The **energy efficiency of a fuel cell is generally between 40 and 60%**; however, if waste heat is captured in a cogeneration scheme, efficiencies of up to **85%** can be obtained.

Fuel cells efficiently convert diverse fuels directly into electricity without combustion, and they are key elements of a broad portfolio for building a **competitive, secure, and sustainable clean energy economy.**

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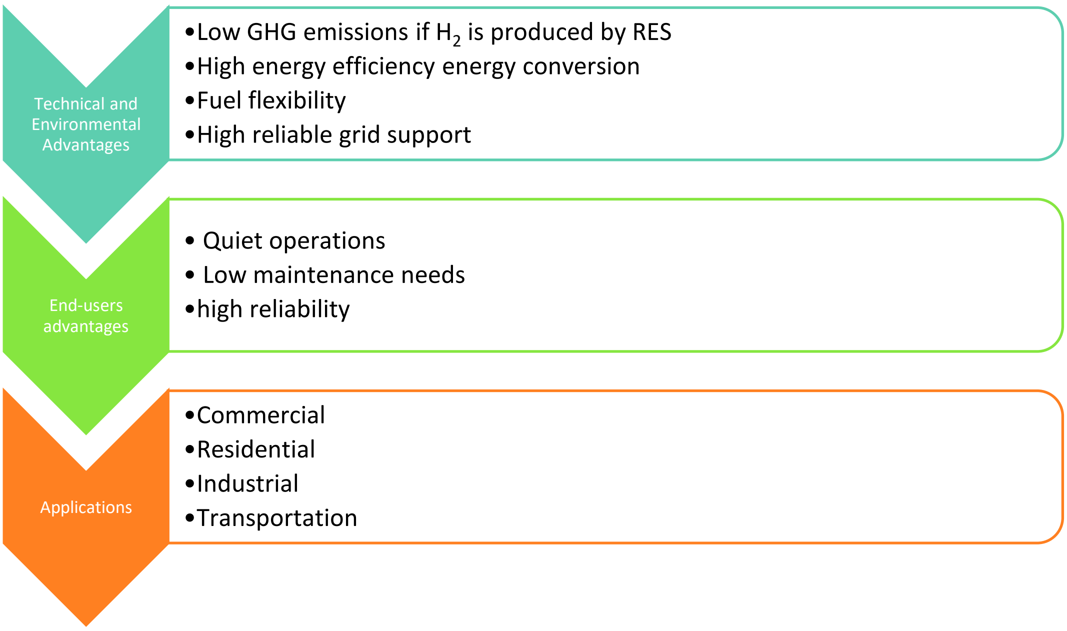

Technical and Environmental Advantages

- Low GHG emissions if H_2 is produced by RES
- High energy efficiency energy conversion
- Fuel flexibility
- High reliable grid support

End-users advantages

- Quiet operations
- Low maintenance needs
- high reliability

Applications

- Commercial
- Residential
- Industrial
- Transportation

Figure 4 General advantages and applications for fuel cells

3.1 Components of a PEMFC [7]

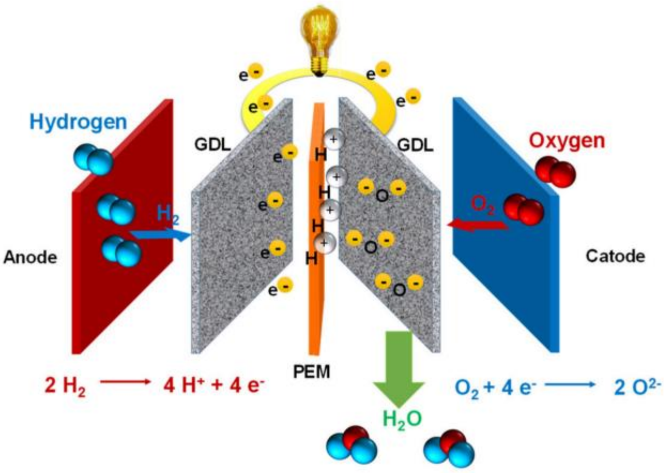

Figure 5 Schematic diagram showing the components of a single PEMFC

The primary components of a PEMFC are:

- the proton exchange membrane as solid electrolyte
- an electrically conductive, porous gas diffusion layer (GDL) with the Microporous Layer (MPL)
- an electro-catalyst of electrically conducting material (forming the electrodes) at the interface. The electrodes comprise the anodic and cathodic Catalyst Layer (CL), the Microporous Layer (MPL), and a Gas Diffusion Layer (GDL). PEMFC technology uses platinum as catalyst. Platinum catalyst constitutes one of the largest cost components in the fuel cells.

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Generally, the three components are joined together to form a Membrane Electrode Assembly (MEA) which is the heart of the PEMFC.

The MEA is placed between two Bipolar Plates (BP), where gas flow channels (GFC) are grooved or placed. These also provide electrical interconnection.

The hydrogen oxidation reaction (HOR) and oxygen reduction reaction (ORR) take place at the triple-phase boundaries in the catalyst layer of anode and cathode, respectively. The CL also provides pathways for both reactant transport and electron/proton conduction.

The polymeric membrane constituting the electrolyte provides proton conduction, electronic insulation, and separation of reactant gases [6]. The membrane should be durable, robust and resistant to chemical attack. The choice of membrane materials depends on the temperature range at which the fuel cells are operating so that the membrane should have a wide operating temperature range -30°C to 200°C. Normally for PEMFCs which operate at temperatures below 100 °C, **sulfonated polymers are the most used material. They are classified as ionomers as some of the repeat units constituting the polymeric chain are ionized**.

The GDL and MPL placed between the CL and BP conduct electrons and heat, transport gas reactants, and enable water management.

Bipolar Plates are made of e.g. graphite with grooved gas flow channels (GFCs). Having high corrosion resistance and electrical conductivity, graphite possesses large gas permeability and is brittle, making it difficult for mass production and long-term use. Several BPP materials have been explored for commercial PEMFCs, including carbon composites, aluminium, stainless steel (stamped and laser welded in the form of metal sheets for BPPs), and titanium.

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3.2 Components of a SOFC [2]

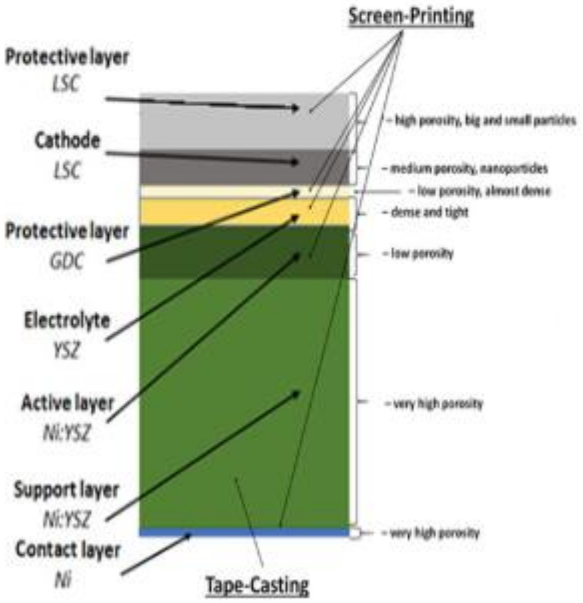

Figure 6 Schematic diagram showing the components, materials and manufacturing methods of an Elcocell^{TM} SOFC

The SOFCs are solid state fuel cells that use an electrolyte of zirconia stabilised with the addition of a small percentage of yttria (Y_2O_3). Above a temperature of about 800°C, zirconia becomes a conductor of oxygen ions (O=). Typically, zirconia-based SOFC operates between 800 and 1100°C. This is the highest operating temperature of all fuel cells. The anode (i.e., the fuel/steam electrode) of the SOFC is usually a zirconia cermet (an intimate mixture of ceramic and metal). The metallic component is nickel, chosen amongst other things because of its high electronic conductivity and stability under chemically reducing conditions. Indeed, during operation the cermet undergoes reduction from NiO-YSZ to NI-YSZ. The zirconia serves to inhibit sintering of the metal particles and provides a thermal expansion coefficient comparable to that of the electrolyte so to ensure thermal and mechanical stability of the cell at all temperatures. The thick substrate is also a cermet connecting to the nickel contact layer which connects to the bipolar plates.

Most SOFC cathodes (i.e., the oxygen electrode) are now made from electronically conducting oxides or mixed electronically conducting and ion-conducting ceramics, such as strontium-doped Lanthanum Manganite/Iron/Cobaltite (LSM/LSCF/LSC), which have a perovskite structure. Their porous texture allows rapid mass transport of reactant and product gases.

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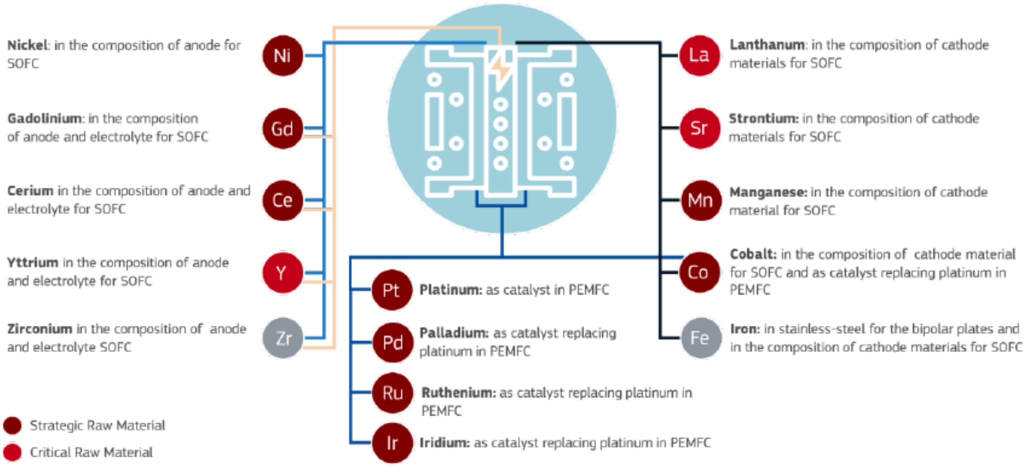
4 Valuable materials in PEMFCs and SOFCs [3]

FC and hydrogen storage technology production requires about 24 raw materials. A selection of them is represented below.



Source: JRC analysis.

Figure 7 Selection of raw materials used in fuel cells and their function (from [3])

In summary:

- For PEMFCs, which operate at low temperatures (< 100°C), the most important processed materials are carbon paper/cloth-based or other carbonaceous materials including carbon nanotubes and graphene nanosheets treated with polytetrafluoroethylene (PTFE) for the GDL and platinum (Pt) or platinum alloys for the catalyst. The electrolyte is a membrane made of perfluorosulfonic acid (PFSA) polymers and the bipolar plates are non-porous graphite, coated or non-coated metals and polymer-carbon or polymer-metal composites.
- SOFC requires different processed materials such as nickel-yttria-stabilised zirconia (Ni/YSZ) cermet or nickel-gadolinium doped ceria (Ni-GDC) used as anode materials, while strontium-doped lanthanum manganite (LSM) or strontium-doped lanthanum cobaltite ferrite (LSCF) are used as cathode materials. The electrolyte is YSZ or GDC. High temperature stainless steel is used for the bipolar plates.

Therefore, the most important raw materials used in FCs are platinum and platinum group metals (PGM), graphite, yttrium, zirconium, cerium, gadolinium, lanthanum, cobalt,

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manganese, iron ore and nickel. The majority of these materials (about 18 of the 24) are deemed critical by the EU according to the 2023 Critical Raw Materials list [4][2].

The EU is almost entirely dependent on foreign sources. China is the world's leading producer of several CRMs, producing 100% of the total output for cerium and yttrium, more than 85% of the total output for lanthanum and gadolinium and 65% of natural graphite. Cobalt world production is largely sourced from the Democratic Republic of the Congo (63%) and processed in China (63%), while PGMs mostly come from South Africa. Most of these countries have low environmental standards and poor social governance. In particular, platinum mining is mainly concentrated in South Africa (71%), followed by Russia (12%) and Zimbabwe (8%). This fact, combined with competing demand from automotive catalytic convertors, chemicals production, medical services and electronic appliances, keeps its price high.

As demand for hydrogen technology increases, so will demand for such materials, and this poses the hydrogen technologies supply chain at risk. Other supply chains, key to the economic and decarbonized growth of Europe, face the same challenges. For this reason, in March 2023, the EU Commission issued the Critical Raw Materials Act [5] as a first set of actions to ensure the EU's access to a secure, diversified, affordable and sustainable supply of critical raw materials.

The Act highlights the need to increase the diversification of materials supply, the improvement of the EU manufacturing opportunities, as well as the need to boost recycling, reuse and substitution of materials. Indeed, finding equally efficient and available alternative materials to platinum as catalyst is not realistic, **thereby recovery and recycling PGMs appears to be the best sustainable and economic solution**. Efforts should focus on R&D to improve the efficiency of recycling technology and the cost of recycling.

Indeed, recovery of materials from End-of-Life hydrogen devices or even from production scraps has the potential for supporting the EU economy, as well as ensuring sustainable

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[2] A criticality approach is one that considers the hazards involved in the creation, utilization, or end-of-life management of a raw material. A raw material is considered critical when:

- It is utilized in industries that are considered strategic for the government or the economy (transports, energy, defense, and electronics);
- It is difficult to substitute in the near future;
- Its reserves and production are regionally concentrated;
- It has a wide range of industrial applications with significant economic value.

management of waste arisings. However, according to the findings of previous research initiatives, there are very few examples of viable and up-scaled technologies for the recovery and recycling of critical raw materials from fuel cells (FCs) and electrolysers.

BEST4Hy overall objective is to bring recycling technologies to TRL5, adapted specifically for PEMFCs and SOFCs, which would ensure the maximization of recycling of critical raw materials including Platinum Group Metals (PGMs), rare earth elements, cobalt and nickel. The End of Life (EoL) strategy supported is accompanied by LCC and LCA evaluations to ensure it delivers the best (cost effective and low environmental impact) material for closed loop and open loop recycling. Materials are evaluated for quality and performance in remanufactured PEMFCs & stacks and SOFCs, so to deliver a concrete validation of the circularity potential within the hydrogen device industry.

Environmentally sound product design (eco-design) can also play a big role in making products more energy and material efficient. The EU Eco-design Directive [6], which has been in force since 2009, is underpinned by the notion of environmentally sound product design. It states the minimum requirements products must meet drawing up product-specific regulations. These stipulate binding minimum requirements for environmentally sound product design. At present, 22 product groups are subject to such implementing regulations. The EU is rethinking its approach on sustainability of products in general, starting from the experience gained with the 2009/125/EC Directive.

With the launch of the initiative, the “Eco-design requirements for sustainable products”, a proposal for a new Eco-design for Sustainable Products Regulation (ESPR) was published in 2022.

The proposal establishes a framework for setting eco-design requirements for specific product groups to significantly improve their circularity, energy performance and other environmental sustainability aspects, so to promote more sustainability over the whole life cycle of goods placed in the market. This could also increase recycling output in the near term, boost demand for recycled goods, increase their recycled content, and increase recycling rates in the EU.

Project eGhost is focusing on eco-design of hydrogen devices. More information can be found at this link: https://eghost.eu/

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[5] COM(2023) 160 - Proposal for a regulation of the European Parliament and of the Council establishing a framework for ensuring a secure and sustainable supply of critical raw materials

[6] Directive 2009/125/EC of the European Parliament and of the Council of 21 October 2009 establishing a framework for the setting of Eco Design requirements for energy-related products

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