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

# TRAINING MODULE 2
# Chapter_2

## Description and manual to perform the existing and optimized recovery & recycling technologies (PEMFC)

**Document Details**

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| Prepared by | ENVI |
| Input from | IDOLab |

**Document Details**

x PU - Public

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

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# Abbreviations

| | |
|---|---|
| CEA | CEA research centre |
| CCM | Catalyst Coated Membrane |
| CL | Catalyst Layer |
| FC | Fuel cell |
| GDL | Gas Diffusion Layer |
| HRD | Hensel Recycling |
| IDO | IDO-LabL |
| MEA | Membrane electrode assemblies |
| PEM | Proton Exchange Membrane |
| Pt | Platinum |

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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: Recycling technologies
- Module 3: Technical results and economical aspects
- Module 4: Measures towards the take up.

Module 2 is dedicated to the recycling technologies, providing knowledge on the adaptation and designing of existing and novel processes and an overview of the processes developed within the project.

Module 2 should be organized as follow:

- Chapter 1: Introduction to the current existing recovery & recycling technologies
- Chapter 2: Description and manual to perform the existing and optimized recovery & recycling technologies (PEMFC);
- Chapter 3&4: Description and manual to perform the novel recovery & recycling processes developed within the project (PEMFC/SOFC);

Video tutorials on how to perform the recovery and recycling processes above described are also available. For more information, users can consult the associated deliverables describing the activities performed in BEST4Hy:

- D1.2: Technical report on the adaptation of existing technology (hydrometallurgy process) for PEMFC material recovery: results and design
- D1.5: Pilot-scale plant (TRL5) based on 3 recycling technologies for PEMFCs
- D2.3: Report on the evaluation of MEA including recycled materials in a small single cell of PEMFC
- D2.6: Report on the evaluation of MEA including recycled materials in PEMFC stack_PU
- D3.3: Pilot-scale plant (TRL5) based on two integrated existing recycling technologies for SOFCs_PU
- D3.5: Technical report on open loop analysis of different scenarios
- D4.3: Technical report on developed recovery technologies for LSC cathode materials_PU

<u>The present document refers to Chapter 2: Description and manual to perform the existing and optimized recovery & recycling technologies (PEMFC)</u>

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# 2 Introduction

This chapter reports results and achievements linked to the WP1 "Existing and novel Technologies of PEM Recycling: Proof of Concept" with specific focus on the recovery and recycling processes for Platinum from MEAs, after EoL PEMFC disassembling.

Well-established low environmental impact technologies for recovery and recycling of end-of-life PEM fuel cells still do not exist. In BEST4Hy, the Consortium focused on both optimization of existing technologies and exploration of novel technologies for the recovery of critical /precious (PGM)/ rare earth elements materials. The current chapter reports achievements of optimized recovery process at TRL5 for PEMFC and Platinum specifically.

Research activities have been carried out by IDO Lab and Hensel Recycling while EKPO provided the EoL fuel cells. Some MEAs were also provided by the CEA as obtained from their gas dismantling process (under patenting). More details on the topics explained in this chapter are available in the following public deliverables:

- D1.2: Technical report on the adaptation of existing technology (hydrometallurgy process) for PEMFC material recovery: results and design
- D1.5: Pilot-scale plant (TRL5) based on 3 recycling technologies for PEMFCs

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# 3 Hydrometallurgical process to recover Pt-salt

The hydrometallurgical process is currently the reference technology for recovery of Pt-salt from EoL PEMFCs at TRL3 [1].

Within BEST4Hy, operation parameters have been studied and optimised for the scaling-up of the technology up to TRL5.

The main parameters taken into consideration are:

- the %vol oxidants, influencing the leaching and the separation efficiency;
- time of the processes;
- use of a ion exchange resin to optimize Pt separation.

The following scheme has been studied and implemented by IDO-Lab (Figure 1):

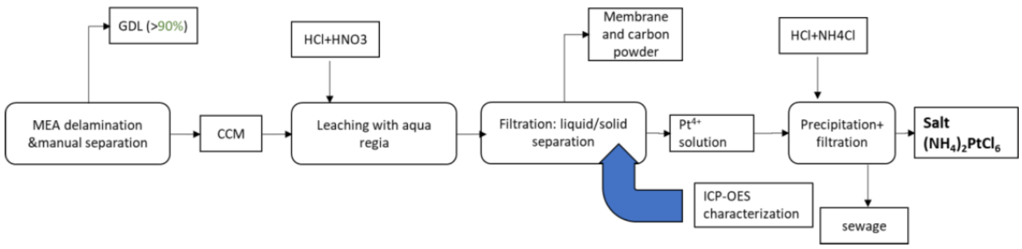


*Figure 1 HMT process at IDO-Lab*

The process has been tested on different MEAs, considering different type of membrane provided by EKPO (surface, operating hours, etc. ) and disassembling process, i.e manual dismantling operated by Hensel Recycling and novel dismantling process performed by CEA.

Process steps are described in the following sections.

## 3.1 Leaching

The leaching agent is a mixture of a strong acid and an oxidant. Aqua regia is typically used for the leaching of spent catalysts in the recovery of PGMs (on carbon support). It is a solution of hydrochloric acid and nitric acid with a molar ratio of 3:1. Its use as a leaching agent allows Pt-recovery percentages above 95%. The leaching process generates very harsh working conditions, due to high acid concentration with pH<1. Although different acid/oxidant solutions are possible, aqua regia has very high recovery efficiency.

**Leaching process**:

- Use of pure CCM, ideal sample size 100g
- Leaching with HCl (36% conc., 150mL) and HNO3 (63% conc., 50mL)
- 1h heating and stirring (nominal power of heating plate: 630W), afterwards 1h stirring
- Temperature = 100° C

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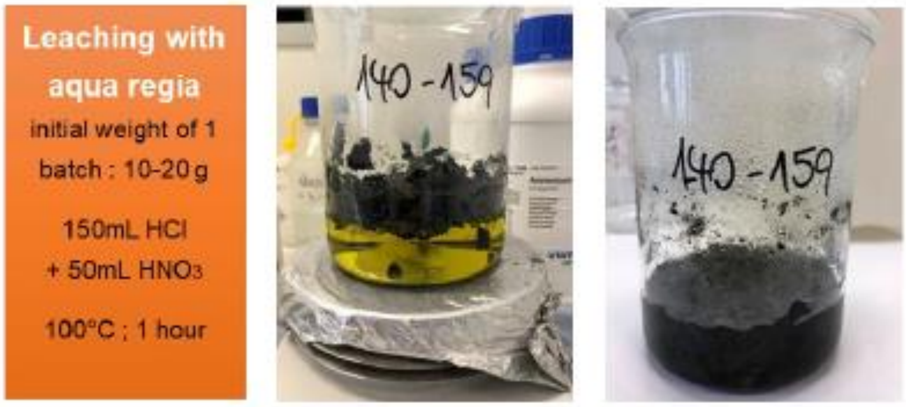


*Figure 2 Leaching with aqua regia step – BEST4Hy tests and samples*

The CCM is dissolved in aqua regia and split in 2 fractions:

- Pt in solution and
- foil containing membrane and carbon

## 3.2 Filtration

After leaching, a filtration stage removes the membrane (ionomer) and carbon particles from the PGM-containing solution

**Filtration steps**:

The liquid is filtrated to obtain a clear Pt solution into a volumetric flask, using blue ribbon filter paper. The Pt concentration dissolved out of CCM after leaching process with aqua regia is around 90-95%. During BEST4Hy project, this was quantified by ICP-OES (ICP nominal power: 1300W, time: 0,5 h; little consumption of Argon gas, Scandium, buffer (NaCl)).


*Figure 3 Solid/liquid separation step - BEST4Hy tests and samples*

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## 3.3 Precipitation and Filtration

In this last step, the Pt-rich stream coming from the separation process is treated with ammonium chloride ($NH_4Cl$) to precipitate Platinum as $(NH_4)_2PtCl_6$, which is filtered and recovered in solid form as the final product of the process.

**Precipitation and filtration steps**:

- The concentrated solution is precipitated with ammonia chloride: $NH_4Cl$ (saturated solution 250g/L) 20mL using a piston pipette. As a result, Pt salt is obtained from the Pt rich solution
- Vacuum filtration using cellulose filter (blue ribbon) paper 2µm to obtain 2 fractions: Pt as Pt salt in solid form and the filtrate (waste solution)
- Pt-salt is dried in the oven before final weighing: drying Pt-salt $(NH_4)_2PtCl_6$ in drying cabinet/furnace (nominal power:1400W, drying time: 24h at 50°C)


*Figure 4 Precipitation step – BEST4Hy tests and samples*

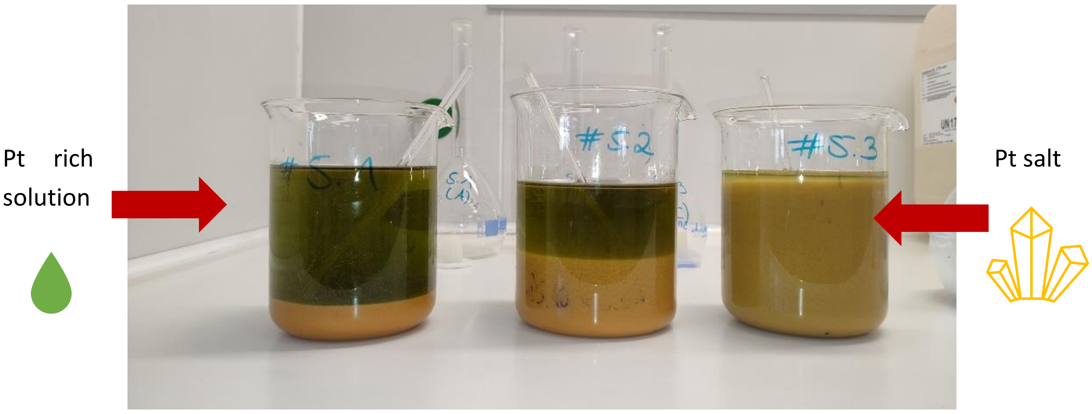


*Figure 5 BEST4Hy samples: Pt rich solution on the left, Pt salt on the right.*

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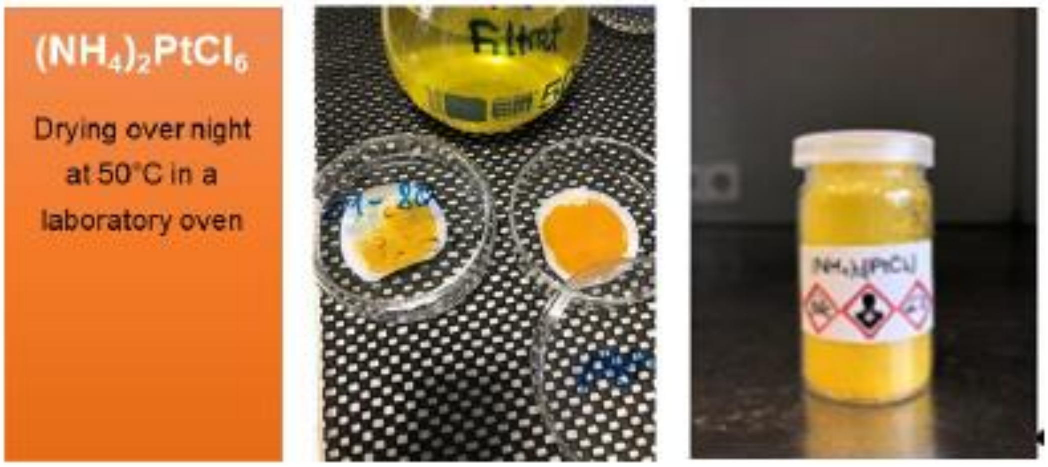


*Figure 6 Filtration step – BEST4Hy tests and samples*

## 3.4 Comparison with hydrometallurgy process from literature

| Main issues during laboratory optimization | before | after |
|---|---|---|
| Batch size<br>before: max. 5 g in flask<br>after: 50-100 g in beaker glass |  |  |
| Filtration: choice of filter paper | white ribbon | blue ribbon |
| Choice of Filtration technique:<br>before: Buchner funnel<br>after: Vacuum Filtration |  |  |
| Vacuum Filtration: mesh size of membrane filter paper | 0,45µm |  0,2µm |

*Figure 7 Partial laboratory parameter optimization in BEST4Hy process*

The average yield increased to more than 90% for the optimized BEST4Hy hydrometallurgy process.

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# 4 References

[1] L. Duclos et al., Process development and optimization for platinum recovery from PEM fuel cell catalyst, Hydrometallurgy Journal, Volume 160, March 2016, Pages 79-89.

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