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

TRAINING MODULE 2

Chapter_3

Description and manual to perform the novel recovery & recycling processes developed within the project (PEMFC)

Document Details

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Document Details

x PU - Public

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

Clean Hydrogen Partnership

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Abbreviations

AB	Advisory Board
AD	Alcohol Dissolution
BPP	Bipolar Plate
EoL	End-of-Life
GA	Grant Agreement
GDE	Gas Diffusion electrode
GDL	Gas Diffusion Layer
GPC	Gel Permeation Chromatography
HMT	Hydrometallurgical Process
HRD	Hensel Recycling Deutschland
HTEL	High Temperature Electrolysis
HTH	Hydrothermal Treatment
MEA	Membrane Electrode Assembly
PFSA	Perfluorosulfonic acid
PEMFC	Polymer Electrolyte Membrane Fuel Cell
PGMs	Platinum Group Metals
US	Ultrasound Bath
WP	Work Package

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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 to 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 3: Description and manual to perform the novel recovery & recycling processes developed within the project (PEMFC).</u>

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2 Introduction

Within project BEST4Hy, existing PGM recovery technologies have been studied and optimised to higher technology level. This chapter reports results and achievements linked to novel recovery processes that have also been investigated, i.e.:

- electroleaching and electrolixiviation processes by the CEA,
- alcohol dissolution process by Hensel Recycling.

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

3 Electroleaching and electrodeposition to recover metallic Pt

3.1 Concept of the process

The process of electro lixiviation/deposition is based on a patented process proposed by CEA institute (EP3263744 A1 2018-01-03).

The proof of concept is demonstrated in a three-electrode cell as presented below.

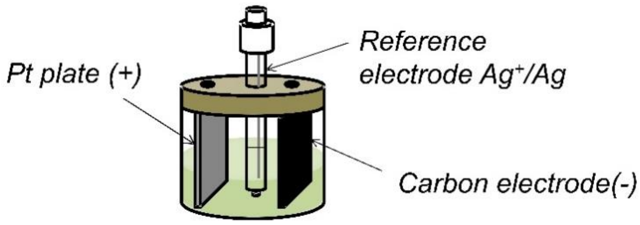

Figure 1 Scheme of the three-electrode cell

In this process, platinum can be recovered from electrochemical leaching in an ionic liquid mixture (IL) containing chloride ions. The ionic liquids selected for the process are:

- 1-Butyl-3-methylimidazolium bis trifluoromethyl sulfonyl imide (BMIM TFSI);
- 1-Butyl-3-methylimidazolium chloride (BMIM Cl).

Figure 2 shows the respective molecular structures of the two selected ionic liquids.

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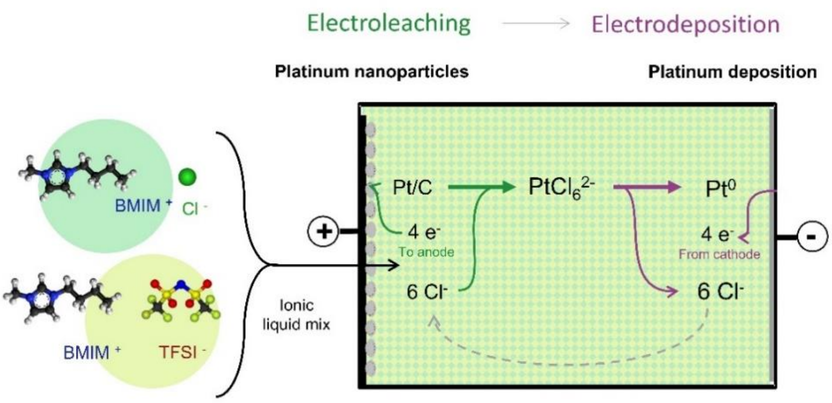

Figure 2 Scheme of simplified electrochemical leaching and deposition process operating mode

The Pt nanoparticles are electrochemically leached at the anode, forming platinum and chloride complexes, and zero-valent platinum is electrochemically deposited at the cathode of the same electrochemical cell.

The electro lixiviation/deposition is a user-friendly and efficient process for the following reasons:

- no use of organic solvents nor acids;
- no toxic gas emissions;
- only one step process.

3.2 Method: Development of a new electrochemical cell

First step is the development of a new electrochemical reactor, where the electro lixiviation and deposition of Pt/C coming from MEA material can happen. The electrochemical reactor needs to be realized with the following general parameters as reference (Figure 3 for an example):

- high electrode surface area;
- small inter-electrode distance;
- temperature controlled.

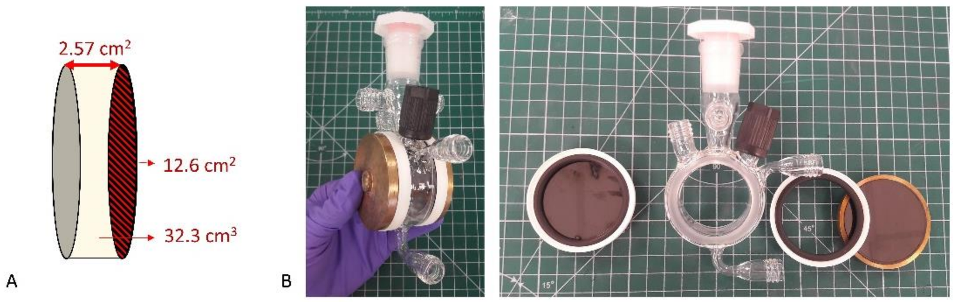

Figure 3 Dimensions of the improved electrochemical vessel (A) and pictures of the vessel (B)

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Process parameters to be controlled for the electro lixiviation process performance in the reactor:

- concentration of chloride anions;
- concentration of leached platinum in the medium;
- operating potential on the three-electrode cell.

The process has been initially tested on one type of membrane electrode assembly (MEA) composed of a gas diffusion electrode (GDE).

On optimisation step was planned during the project to improve and to adapt the process to different kind of MEAs: membrane with a catalyst coated membranes (CCM) where the catalyst layer (CL) is coated on the membrane; membrane with a catalyst coated substrates (CCS), for which the CL is coated on the gas diffusion layer (GDL), forming a gas diffusion electrode (GDE).

After tests, the process efficiency has been analysed by comparing the quantities of Pt electrochemically leached and deposited. The values have been compared with initial material specifications provided by EKPO.

For further information, please consult European Patent EP3263744 A1 2018-01-03.

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4 Alcohol dissolution process for Platinum and Ionomer recovery

4.1 Concept of the process

Conventional technologies for recovery of valuable materials from PEMFCs, such as Hydrometallurgical Treatment (HMT) and High Temperature Treatment (HTH), which is an incineration process, succeed in salvaging Pt, but no other valuable materials as, for instance, the ionomer membrane.

The alternative process of Alcohol Dissolution (AD) enables the recovery of Pt and ionomer.

The AD process developed by HRD and IDO-Lab within project BEST4Hy consists of an alcohol dissolution in a high temperature and pressure autoclave reactor, which is fed with pure CCM in an alcohol/water mixture, followed by different separation steps to split the ionomer solution from Pt/C. The mixture is transferred in a laboratory centrifuge, the ionomer solution is removed by mechanical separation (e.g., decanting) and it is then filtered to guarantee high quality of ionomer - and prevent any particles in the solution.

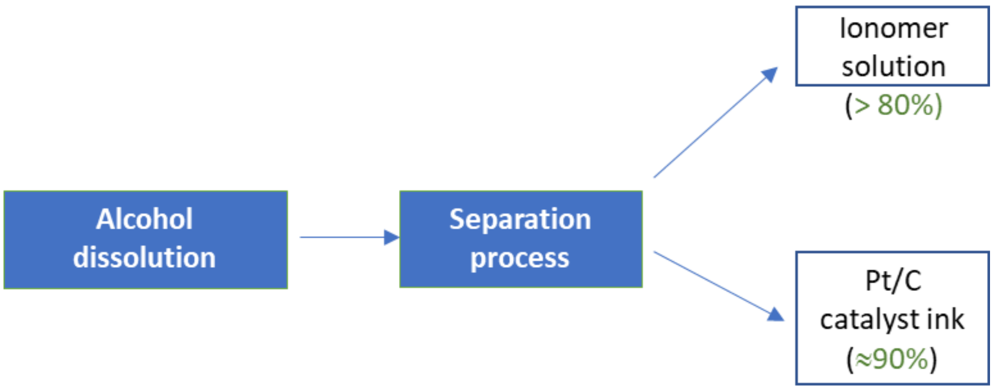

Figure 4: Scheme of IDO-Lab AD process

<u>After the recovery, Ionomer is further analysed and characterized to assess its quality as secondary materials and features for the following reuse, e.g., remanufacturing of new MEA.</u>

4.2 Method: the Alcohol Dissolution process

The process was developed within BEST4Hy by first identifying the best solvent and then the steps required to separate ionomer and the Pt rich component. Samples treated are a mixture of CCM and GDL.:

1. **Alcohol solvent selection**: ethanol has been selected as best solvent for the ionomer dissolution and the separation of Ionomer by Pt ink. Ethanol is a low cost- and low-environmental impact substance, preferred to other competitive solvent such as methanol.

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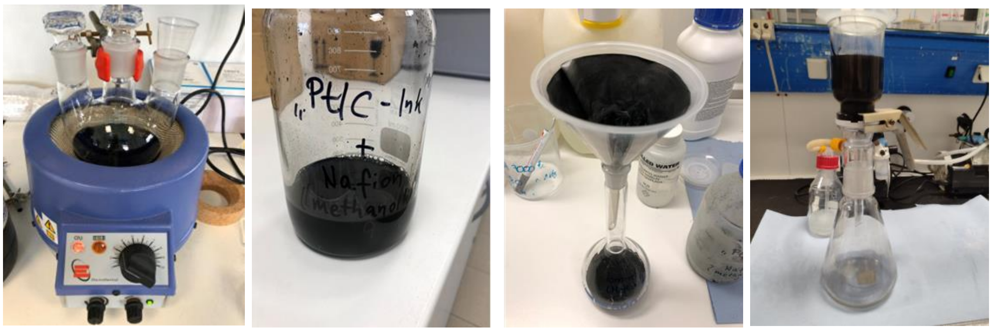

Figure 5 Dissolution tests with different alcoholic solvents

2. **Pressure autoclave reactor**: literature refers of better dissolution performance at high pressure and temperature conditions. Within BEST4Hy, the AD process was performed in a pressure vessel. Alcohol/water mixture is used as solvent solution.

Figure 6 Dissolution tests using pressure vessels.

3. **Pre-filtration**: a centrifugation process is needed to allow the separation of the coarse part so that clogging is prevented during secondary filtration.

4. **Mechanical separation**: decanting allows the Ionomer deposition within the solution and further separation.

5. **Filtration - Pt/C ink and ionomer solution separation**: vacuum filtration step using cellulose filter papers with 0,45 µm.

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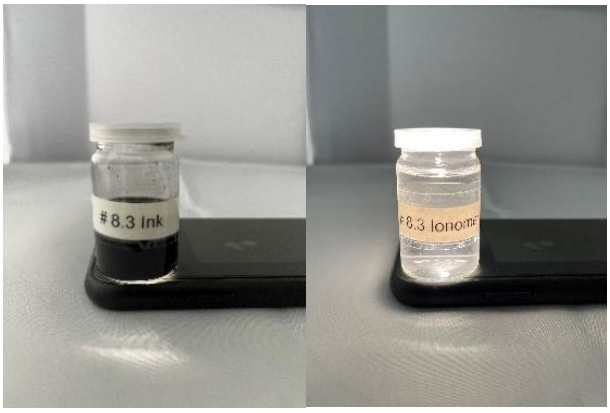

Figure 7 Example of separated fraction, Ionomer and Pt ink

No more detailed information can be specified due to the existence of a Patent procedure ongoing for the present technology.

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