

Cost-effective PROton Exchange MEmbrane WaTer Electrolyser for Efficient and Sustainable Power-to-H2 Technology

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Table of Content

Document hi	story2
List of Figure	s5
List of Tables	۶6
Executive Su	ımmary7
1	Introduction
2	MEA based on Non-PGM anode electrocatalyst for testing in PEM water electrolysis
single cell	9
2.1	Electrochemical investigations9
2.2	Results9
2.3	Conclusions
3	MEA based on low-PGM anode electrocatalyst for testing in PEM water electrolysis
single cell	
3.1	Electrochemical investigations
3.2	Results
3.3	Conclusion
4	Cross-over, Performance and Stability of a reinforced membrane
5	Assessment of low iridium loaded membrane electrode assemblies
5.1	Performance assessment of iridium supported on antimony tin oxide with an 70wt.%
iridium	21
5.2	Determination of the sheet resistance and in-plane electrical resistivity for the
catalysts stud	died21
5.3	Validation of the produced MEA for the NEL stack23
5.4	Conclusion
6	Recycling of CRMs from MEAs24



List of Figures

Figure 1. Polarisation curves (a) and AC-impedance spectra (inset: high magnification) (b) at 1.8 V and 2 V at 80 °C for the MEA with Ag/Ti-suboxides anode catalyst; Polarisation curves (c) and AC-Figure 2. Steady-state durability test at 0.6 A cm⁻² and 80 °C of the MEA with Ag/Ti-suboxides anode catalyst (a); Polarization (b) and AC-impedance spectra at 1.8 V (c) and 2 V (d) at 80 °C of the MEA with the Ag/Ti-suboxides anode catalyst at BoL and after the steady-state durability test. Figure 3. Voltage cycle test (a) at 1 - 1.8 V and 80 °C and steady-state durability test (b) of the MEA with Aq/Ti-suboxides anode catalyst.....13 Figure 4. Polarisation curves at 80 °C for the MEA with the Ag/Ti-suboxides anode catalyst before Figure 5. j – E_{cell} curves recorded at 80°C and P_{amb} for electrolyzers based on 0.4 mg_{Pt}cm⁻² as Figure 6. Force Vs. Strain Measurements of previous commercial membranes and the newly Figure 7. Current-Voltage curves of previous commercial membranes and the newly developed Figure 8. Performance assessment (a) and cross-over testing (b) results obtained from single-cell Figure 9. Single-cell testing of the synthesized Ir-ATO 70wt.% as MEA with a loading of 0.2 mg/r Figure 10. Assessment of the sheet resistance and in-plane electrical resistivity for the catalyst layers used within this project. The high load benchmark IrO_2 catalyst is shown in (a), and in (b) with the targeted loading of 0.2 mg_{lr} cm⁻², while (c) shows the Ir-ATO 50 wt.% catalyst used in the Figure 11. Comparison between the measured sheet resistance and the obtained HFR from single-Figure 12. Validation of the stack MEA by cutting out a specimen and testing it in a single cell. ... 23 Figure 13. Twelve EoL CCMs (N117, Pt/C, IrO₂) provided by Julich. Electrode area=25cm².......24 Figure 15. MONOLITHOS hydrometallurgical leaching process to recover CRMs from EoL Figure 16. Studying the delamination and separation of the catalyst layers from the MEA to obtain



List of Tables

Table 1. Expansion and Area Resistance of previous commercial membranes and the newly	
developed reinforced membrane	. 18
Table 2. Samples provided by Julich to MONOLITHOS for recycling.	. 24



Executive Summary

The deliverable D2.6 "3rd annual report on MEA performance assessment" provides the main results obtained with the CRM free catalysts and the reduced CRM catalysts-based MEAs as well as catalyst recycling and performance with different membranes. The main findings are the following:

- A non-PGM anode catalyst for PEM electrolysis anodes consisting of Ag/Ti-suboxides showed good stability but to achieve a current density of 2 A/cm² a higher voltage was required compared to PGM based catalysts.
- Anodes containing low PGM loading (0,2 mg/cm² instead of 2 mg/cm²) based on supported Ir catalysts can obtain similar performances than standard anodes based on unsupported Ir catalysts.
- A reinforced membrane was developed and allows the combination of the stability of a thicker membrane and the performance of a thinner membrane.
- A method and sensor to determine lateral conductivity in catalyst layers has been developed and allows showing that lateral conductivity of low loaded supported Ir is similar to that of high loaded unsupported Ir
- 99% of Pt and 60% of Ir can be recovered by an environmentally friendly hydrometallurgical recycling process.



1 Introduction

Water electrolysis is a very promising technology for sustainable hydrogen generation using renewable electrical energy. The excellent performance and dynamic behavior for storing electrical energy in hydrogen allows polymer electrolyte membrane (PEM) electrolysis to cover the gap between the intermittent renewable power production and the grid demand at different time horizons and scales. To allow for a widespread utilisation of the PEM electrolysis technology, a significant reduction of the capital costs is strongly necessary. To achieve such objective, precious metal catalysts used in the electrolysis systems should be minimised.

The main objective of WP2 is to develop advanced, highly efficient and durable membrane electrode assemblies (MEAs) through the integration the novel electro catalysts developed in the WP1, based on reduced CRM or free catalysts, and new durable membranes developed within WP2.

Deliverable D2.6: "3rd annual report on MEA performance assessment" provides the main results obtained with the CRM free catalysts and the reduced CRM catalysts based MEAs in terms of performance and stability obtained in PEM electrolysis single cells. It also contains measurements that help understanding why MEAs with reduced CRM catalysts can obtain similar performance than MEAs with high CRMs. The influence of new membranes is shown as well as the recycling process that can help reducing total cost of ownership by recovering precious metals at the end of service life.



2 MEA based on Non-PGM anode electrocatalyst for testing in PEM water electrolysis single cell

A membrane – electrode assembly (MEA), consisting of the anode Ag/Ti_nO_{2n-1} and conventional Pt/C cathode catalysts deposited on a 212 NAFION® membrane (thickness 50 μ m), was investigated at CNR to assess the performance and durability of the PGM free oxygen evolution catalyst in an acidic environment. A promising performance, of 0.6 A·cm⁻² at 2 V/cell at 80°C, and an excellent stability (degradation rate < 14 μ V/h during a 1000 h test) were achieved for the electrolysis cell based on a cost-effective metal anode electrocatalyst.

2.1 Electrochemical investigations

The performance and stability of the PEM electrolysis cell were investigated at CNR by means of electrochemical studies at 80° C and under ambient pressure conditions. Deionized water (Milli – Q integral Millipore) with a resistivity of 18.2 m Ω ·cm, was fed to the anodic compartment with a flow rate of 1 ml·min⁻¹cm⁻² and was left to recirculate at the same temperature of the cell. Galvanostatic polarization curves and durability tests (cell potential *vs.* cell current and cell potential *vs.* time, respectively) were obtained by using a TDK GenesysTM 25400 – MD – 3P400 power supplier. A cut off of 2.5 V was selected.

Electrochemical impedance spectroscopy (EIS) analyses were performed at 1.5 V and 1.8 V by varying the frequency from 100 kHz to 100 mHz in single sine mode by using an Autolab PGSTAT 302 Potentiostat/Galvanostat, equipped with a Metrohm 20 A booster and a frequency response analyser (FRA). The amplitude of the sinusoidal excitation signal was 0.01 V r.m.s. 50 log scale frequencies were swiped in descending order. The series resistance was determined from the high frequency intercept on the real axis in the Nyquist plot.

2.2 Results

Regarding the electrochemical characterization, polarization curves (cell potential as a function of current density) and electrochemical impedance spectroscopy (EIS) were carried out (Fig. 1a-b). The Ag/Ti-suboxides catalyst (30-70 at %) thermally reduced in a H_2 - N_2 stream showed an ohmic loss corrected (IR-free) single cell performance of 0.6 A cm⁻² at 2 V and 2 A cm⁻² at 2.15 V (Fig. 1a). The raw performance at 0.6 A cm⁻² was 2.1 V/cell. This corresponded to a voltage efficiency of 70%. The cell voltage at 0.6 A cm⁻² is assumed as key performance indicator of the voltage efficiency since



this is a typical operating current density of advanced liquid alkaline electrolysers that do not make use of PGM metals.

The electrochemical characterization of a single cell based on IrO₂ as anode and Pt/C as cathode is reported in Fig. 1c, 1d. The single cell voltage of the MEA with Ag/Ti-suboxides anode catalyst was higher than a conventional PEMWE based on a precious Ir-based anode (Fig. 1a, 1b vs. Fig. 1c, 1d). The total anode catalyst loading was significantly higher in this work compared to conventional PEM electrolyser (12 vs conventional 2-3 mg cm⁻² for the benchmark IrOx catalyst. However, the silver loading in the Ag/Ti-suboxides anode catalyst (3.3 mg cm⁻²) was comparable to the standard Ir loading in conventional PEMWE anodes.





The behaviour of the electrolysis cell based on the Ag/Ti-suboxides anode catalyst was studied by impedance spectroscopy at 80 °C and two different cell voltage values, 1.8 and 2 V, representative



of activation and ohmic-mass transfer control regions, respectively (Fig. 1b). Similar series resistance values (intercept of high frequency on the real axis) were recorded by Nyquist plots, 145 m Ω cm², for the two different cell voltages (Fig. 1b).

The membrane usually gives the main contribution to the series resistance, whereas the polarization resistance (Rp) is mainly due to charge and mass transfer. The Rp for the two voltage values, 1.8 and 2 V, decreased significantly as the cell voltage value increased (Fig. 1b). The Rp values at 1.8 V and 2 V were 1.48 Ω cm² and 0.210 Ω cm², respectively. Rp was estimated from the difference of the low and high frequency intercept of the Nyquist plot on the real axis. The strong decrease of polarization resistance passing from 1.8 to 2 V (Fig. 1b) was largely indicative of an activation control associated to the oxygen evolution at the Ag/Ti-suboxides catalyst. Whereas the Pt/C cathode catalyst being associated to a fast electrochemical process had much lower or negligible impact on the impedance plot.



Figure 2. Steady-state durability test at 0.6 A cm⁻² and 80 °C of the MEA with Ag/Ti-suboxides anode catalyst (a); Polarization (b) and AC-impedance spectra at 1.8 V (c) and 2 V (d) at 80 °C of the MEA with the Ag/Ti-suboxides anode catalyst at BoL and after the steady-state durability test.



The stability of the Ag/Ti-suboxides anode catalyst was studied by a steady-state durability test of 1000 h at ambient pressure and 80°C (Fig. 2). The durability test, performed at 0.6 A·cm⁻², is reported in Fig. 3a. This time-study appeared appropriate to get some first insights about the behaviour of the novel anode electro-catalyst used in PEM water electrolysis in order to identify possible causes of degradation. After a rapid slight increase of the cell voltage in the first hours of operation, a stable cell potential of about 2.1 was observed. It is reasonable to consider that sub-oxides tend to revert to stoichiometric oxides at the potentials relevant for the OER, leading to a reduced conductivity. Such increase of resistivity may have caused the observed increase of cell voltage of a few tens of millivolts, in the first hours of operation of the steady-state durability test.

An interruption of the test after 350 h, due to plant maintenance issues, showed some small additional losses without however affecting much the voltage efficiency. The cell voltage was much higher compared to conventional Ir-based MEAs. For the Ag/Ti-suboxides (30:70) based MEA an overall degradation rate of about 14 μ V/h was observed by removing the first 24 h conditioning period, that correspond to < 0.7 %/1000 h.

The MEA based on Ag/Ti-suboxides anode catalyst was subjected to polarization and impedance spectroscopy studies before and after the 1000 h steady-state test (Fig. 2b-d). The comparison of the polarization curves carried out before and after the durability test is reported in Fig. 2b. A better performance (lower cell voltage) for the polarization curve after the durability test is indeed observed. Such evidence indicates that the increase of the cell voltage during the overall durability test (Fig. 2a) was essentially associated to the load cycle at 350 h. The improvement recorded in the polarization curve (Fig. 2b) was probably due to an enhancement of the catalytic activity and to better interfacial characteristics occurring in the activation and ohmic-mass transfer-controlled regions after the durability test. The increase of performance (i.e. the decrease of cell voltage at the same current density) in the polarization curve (Fig. 2b) was moderate in the activation region (35 mV at 0.6 A cm⁻²) and larger at high current density (95 mV at 1.8 A cm⁻²).

AC-impedance analysis revealed that, after the durability test, a lower Rs was recorded in the impedance spectra at both 1.8 V and 2 V (Fig. 2c, 2d). This was probably due to an in situ purification of the membrane or it was slight thinning of the membrane with time. The Rs decreased by 45 m Ω cm², for the two different cell voltages 1.8 and 2 V (from about 0.15 to 0.1 Ω cm²).



A decrease of polarization resistance at low frequency was evident in the impedance spectra at 1.8 V after the durability test (Fig. 2c) and indicated better interfacial characteristics, between the catalysts and membrane. The Rp decreased from 1.48 to 0.9 Ω cm² at 1.8 V after the 1000 h steady-state durability test (Fig. 2c).

The larger impact on the decrease of the cell voltage of high current density was in part due to the decrease of the series resistance. Anyhow, these results show good catalyst stability properties.



Figure 3. Voltage cycle test (a) at 1 - 1.8 V and 80 °C and steady-state durability test (b) of the MEA with Ag/Tisuboxides anode catalyst. To understand the effect of load cycles, a voltage cycle test was carried out. The same MEA was subjected to a voltage cycle test at 1 – 1.8 V at a low frequency (0.02 Hz) in order to evaluate the single cell behaviour under stress conditions; the duration of this specific voltage cycling was 24 h at 80°C (Fig. 3a). Some current decay during the first hours was observed. The current value recorded at the beginning of the cycle test corresponded to that observed in the polarization curve after the steady-state durability test (Fig. 2a). The initial value was about 70 mA cm⁻² at 1.8 V. The current



decay led to a current of 20 mA cm⁻² after 24 h test. This loss was probably due to the catalyst degradation or change on the anode catalyst oxidation state during this stress test.

Thereafter a steady-state durability test at $0.6 \text{ A} \cdot \text{cm}^{-2}$ and $80 \,^{\circ}\text{C}$ was carried out to evaluate the effect of the voltage cycle test on the cell stability. A comparison on the two different steady state tests, before and after cycle test, is reported in fig. 3b. An irreversible loss of performance was evident but no relevant impact was observed on the steady state degradation rate. A decay of 50 mV was recorded after the 40 h duty cycle test.



Figure 4. Polarisation curves at 80 °C for the MEA with the Ag/Ti-suboxides anode catalyst before and after stability and cycle tests.

Fig. 4 shows a comparison of the polarization curves for the BoL MEA and the MEA subjected to steady-state operation (1000 h) and to the voltage cycles test (24 h). An increase of the cell voltage at very low current was observed after the voltage cycle test procedure. The cycle test also caused an increase of cell voltage in the current density region up to 0.5 A cm⁻² revealing same additional activation losses compared to the MEA not subjected to duty cycles.

2.3 Conclusions

In this regard, the present work provides an electrochemical single cell assessment of a non-PGM anode catalyst for PEM electrolysis applications. The MEA, consisting of Ag/Ti-suboxides anode, Pt/C cathode catalysts and 212 NAFION® membrane, showed promising performance and durability



characteristics in comparison to analogues alkaline systems based on PGM-free catalysts. At 80°C, performances, of 0.6 A·cm⁻² at 2 V and 2 A·cm⁻² at 2.2 V IR-free were observed together with an elevated stability during steady-state operation.

Such electrochemical results clearly show a lower performance for the novel Ag/TiOx formulation compared to a well optimised IrOx-based anode catalyst in a PEM electrolysis cell. However, the performance achieved with the Ag/TiOx anode appears already competitive to liquid alkaline electrolysis technologies.

Some degradation was observed upon application of duty cycles. This had a negative impact on the catalyst behaviour causing some activation losses.



3 MEA based on low-PGM anode electrocatalyst for testing in PEM water electrolysis single cell

A membrane – electrode assembly (MEA), consisting of the anode Ir/ATO (50%) and conventional Pt/C cathode catalysts deposited on a 117 NAFION® membrane (thickness 178 µm) was investigated at DLR to assess the performance of the low PGM content oxygen evolution catalyst in an acidic environment with stainless steel (SS) porous transport layers (PTLs) coated with a macroporous layer (MPL) developed in WP3. A promising performance has been obtained for the use of this MEA which will be used in the short stack of the project.

3.1 Electrochemical investigations

The PEMWE were carried out in a home-made cell setup. MEA consists in NAFION® 117 (178 µm) membrane with Pt/C on the cathode and Ir/ATO (50%) on the anode. On the anode side a MPL-SS PTL with Nb/Ti and a thin Ir or Pt layer coating was used. On the cathode side, a Nb/Ti-SS PTL was employed and a carbon paper sheet (Spectracarb 2050A-1050) was used as an additional layer contacting the cathode catalyst layer. On both the anode and cathode side Ti-BPPs were employed. The cell active area was 4 cm2 and in both setups and tests were carried out at 80°C and ambient pressure. The polarization curves were measured galvanostatically according to the JRC EU-harmonized procedure.

3.2 Results

Regarding the electrochemical characterization, polarization curves (cell potential as a function of current density) are shown in Fig 5.

The results obtained previously for MEAs provided with Nb/Ti-SS PTLs at the anode showed performances similar to those obtained with Ti PTLs for anodes with high Ir loading (2-3 mgIrcm⁻²). MEAs with low Ir loading (0.2 mgIrcm⁻²) have been studied at DLR and the results have shown how a thin layer of Pt or Ir (0.025 mgPGMcm⁻²) on the MPL assumes $E_{cell} = 1.9$ V at a nominal current density of 2 Acm⁻². This results in a performance improvement > 200 mV.





Figure 5. j – E_{cell} curves recorded at 80°C and P_{amb} for electrolyzers based on 0.4 mg_{Pt}cm⁻² as cathode and 0.2 mg_{Ir}cm⁻² as with different MPL-SS PTLs. The performance shown by the Pt-Nb/Ti-SS PTLs as anode for MEAs with low Ir loading have shown to be a strong candidate for the stack in accordance with the project objectives.

3.3 Conclusion

In this regard, a performance of 1.9 V at 2 Acm⁻² has been demonstrated with MEAs with very low PGM charge. This supposes a reduction in the costs in the production of PEMWE cells. For this reason, these MEAs have been developed for assembly in the short stack of the project.



4 Cross-over, Performance and Stability of a reinforced membrane

At Chemours a reinforced membrane type NDP8007 was developed in order to allow using a relatively thin membrane without stability problems. As shown in Table 1 the reinforcement leads to an extremely low lateral expansion while at the same time showing a low area resistance.

Mombrana	X Expansion	YExpansion	Z Swelling	Areal Resistance	
wemprane	(% change) ^a	(% change) ^a	(% change) ^a	(mΩ*cm) ^b	
N115	11	19	21	121	
NDP-8007	< 1	< 1	45	64	
NR212	19	19	19	44	

 Table 1. Expansion and Area Resistance of previous commercial membranes and the newly developed reinforced membrane



Figure 6. Force Vs. Strain Measurements of previous commercial membranes and the newly developed reinforced membrane



In Figure 6 it is shown that the reinforced membrane can sustain a significantly higher force than the Nafion 212 membrane while undergoing only very low deformation. In Figure 7 it can be seen that during measurements at Chemours the performance of NDP8007 is very similar to that of the thin and instable NR212 membrane. By combining these properties NDP8007 should enable a 9% higher efficiency for stacks of the same size or a potential for 60% reduction of active area including a corresponding reduction in catalyst required.



Figure 7. Current-Voltage curves of previous commercial membranes and the newly developed reinforced membrane

The reinforced membrane NDP8007 possessed a measured thickness of $(111 \pm 4 \mu m)$ and a specified thickness of around 100 μ m. Cross-over measurements were performed as before in this project. The MEA used as anode iridium oxide with a loading of 2.2 mg_{lr} cm⁻² and 25 wt% Nafion ionomer content. The cathode possessed a loading of 0.8 mg_{Pt} cm⁻² using 60 wt.% platinum on carbon catalyst and 20 wt.% Nafion ionomer. The electrodes were coated onto the membrane using hot-pressing at 150°C for 3 min, after heating the sample for 13 min before.

Performance and cross-over testing, conducted at FZ Jülich at 80°C with a water flow rate of 50 mL min⁻¹, are shown in Figure 8. The grey lines represent other commonly used membranes or previous results. The studied NDP8007 membrane showed a performance which can be explained by its



thickness and corresponding ohmic resistance. At 1.8 V the current density was around 2.5 A cm⁻² and at 2V around 4 A cm⁻². The cross-over measurement showed that the reinforced membrane NDP8007 obtained higher hydrogen cross-over rates than the thinner membrane NDP8003, but its values remained below the thinnest membrane (NR212) at higher current densities > 1 A cm⁻². The increased cross-over rates compared to the thinner membrane NDP8003 could emerge from the reinforcement.



Figure 8. Performance assessment (a) and cross-over testing (b) results obtained from single-cell testing.



5 Assessment of low iridium loaded membrane electrode assemblies

5.1 Performance assessment of iridium supported on antimony tin oxide with an 70wt.% iridium

Ir-ATO with 50 wt.% iridium showed a performance close to the performance of the hig- loaded benchmark iridium oxide MEA. To test if the performance can be further improved the iridium content of Ir-ATO was increased to 70 wt.% and compared to Ir-ATO 50 wt.% as well as to IrO₂ with a loading of 0.2 mg_{Ir} cm⁻². However, this Ir-ATO 70 wt.% obtained a reduced performance, emerging from the increased high frequency resistance. A deteriorated contact between PTL and the anode catalyst layer because of the reduced thickness of the catalyst layer could be the reason for this behavior. Thus, Ir-ATO remained the best choice for the PEM stack developed within this project.



Figure 9. Single-cell testing of the synthesized Ir-ATO 70wt.% as MEA with a loading of 0.2 mg/r cm⁻².

5.2 Determination of the sheet resistance and in-plane electrical resistivity for the catalysts studied

Lowering the PGM loading while maintaining performance and durability is a major objective of this project. However, the clear assessment by only single-cell testing is sometimes blurred, because many impact factors contribute to the measured performance. Some of these factors amongst others are the single-cell design, used PTL, the gas diffusion layer (GDL) or the electrochemical testing procedure. For this reason, the sheet resistance was determined by a validated method which is currently undergoing the publication process. After determining the sheet resistance, the in-plane



electrical resistivity and in-plane electrical conductivity were calculated. The results for the catalyst layer studied are presented in Figure 10. All shown catalyst layers are deposited on an N117 membrane and measured at 25°C and 25 % relative humidity. The sheet resistance was determined at two compression forces, 1.00 kN and 2.25 kN



Figure 10. Assessment of the sheet resistance and in-plane electrical resistivity for the catalyst layers used within this project. The high load benchmark IrO₂ catalyst is shown in (a), and in (b) with the targeted loading of 0.2 mg_{Ir} cm⁻², while (c) shows the Ir-ATO 50 wt.% catalyst used in the stack.

Comparing the sheet resistances of the different catalyst layers shows that the benchmark catalyst showed at 2.25 kN the lowest sheet resistance of 412 Ω . However, reducing the loading from 2.2 mg_{Ir} cm⁻² to 0.2 mg_{Ir} cm⁻² increased the sheet resistance to 21456 ± 864 Ω , which is nearly 50 times higher than the high-loaded catalyst layer. Instead using a loading of 0.2 mg_{Ir} cm⁻² and iridium supported on antimony tin oxide obtained a sheet resistance of 1247 ± 266 Ω , which is only 3 times higher than the 2.2 mg_{Ir} cm⁻² benchmark catalyst layer. Comparing these results with the HFR recorded during single-cell testing shows that the lower the sheet resistance the lower the obtained HFR, Figure 11.







5.3 Validation of the produced MEA for the NEL stack

The spray-coating was used to validate the MEA performance. For this reason, a produced stack MEA was used which showed a local stain. The tested MEA was cut out without any defects visible. Single-cell testing showed a gap in the performance between the MEA tested in November 2021 and November 2022. Another test was carried out using the benchmark MEA architecture. This test showed the same gap in performance as the tested stack MEA. That led to the conclusion that different batches of materials used (membrane, PTL, etc.) caused this performance reduction. However, the performance of 2 A cm⁻² at 1.9V still fulfilling the project objectives specified.



Figure 12. Validation of the stack MEA by cutting out a specimen and testing it in a single cell.

5.4 Conclusion

The ongoing characterization of the materials showed that the chosen Ir-ATO 50 wt.% is still a promising candidate to overcome performance limitations while using lowered PGM loadings. The validation of the stack MEA showed a performance aligned with the project objectives.



6 Recycling of CRMs from MEAs

MONOLITHOS is responsible for the recycling of CRMs (Critical Raw Materials) from the electrocatalyst powder. The low cost and environmentally friendly hydrometallurgical leaching process for PGMs (Platinum Group Metals) recovery from spent automotive catalyst powders and filters of light and heavy-duty vehicles developed by MONOLITHOS [1] has been applied to study Pt and Ir recovery from End-of-Life MEAs provided by Julich (Figure 13, Table 2). One of the MEAs (MEA20201013_01NU) was only assembled inside a cell, but not measured (pristine). The rest samples have been measured in a single-cell. In addition, one of the MEAs was stored at high humidity (avoid shrinking of the membrane after testing).



Figure 13. Twelve EoL CCMs (N117, Pt/C, IrO₂) provided by Julich. Electrode area=25cm².

MEA code	Anode loading [mglr cm ⁻²]	Nafion content [%]	Pt loading [mgPt cm ⁻²]	Nafion content [%]	lr [mg]	Pt [mg]
MEA20180404_04DH	2,4	25	0,75	20	60	18,75
MEA20180404_01DH	2,48	25	0,73	20	62	18,25
MEA20180226_14DH	2,4	25	0,79	20	60	19,75
MEA20180404_03DH	2,59	25	0,77	20	64,75	19,25
MEA20171120_06DH	2	25	0,76	20	50	19
MEA20151105_03DH	2,19	25	0,84	20	54,75	21
MEA20151105_04DH	2,33	25	0,78	20	58,25	19,5
MEA20151105_05DH	2,4	25	0,84	20	60	21
MEA20150914_02DH	2,19	25	0,82	20	54,75	20,5
MEA20150914_03DH	2,2	25	0,8	20	55	20
MEA20151105_02DH	2,28	25	0,84	20	57	21
MEA20201013_01NU	2,01	25	0,8	20	50,25	20

Table 2. Samples provided by Julich to MONOLITHOS for recycling



MONOLITHOS is equipped with double layer chemical reactors (0,5-150 lt, 1 m³) and large-scale equipment (such as filtration system, vacuum pumps, rotary evaporator, drying furnace and fume hood) that are used for the recovery of PGMs (Figure 14). XRF and ICP-OES analysis are employed to calculate the recovery rate of the metals.



0,5-150 | **double layered chemical reactors** for large scale electrocatalysts synthesis.



Rotary evaporator



Large scale equipment (e.g. vacuum pumps)



Glass Nutsche filtration system



Fume hood for process upscaling





High temperature furnace

1m³ reactor system with refrigerating heater circulator

Figure 14. MONOLITHOS capabilities and premises.

MONOLITHOS studied the recovery of Pt and Ir from the received CCMs using its low cost and environmentally friendly hydrometallurgical leaching process. As mentioned before (Deliverable 2.3), in order to increase the metal nanoparticles that are accessible to the leaching solution, before leaching process, the CCMs are immersed in an alcohol/water mixture to facilitate the separation of the catalyst layers from the Nafion membrane. Different experimental conditions during membrane-electrodes delamination (such as the type of the solvents, solvents' ratio and sonication time) are tuned to achieve the highest recovery of the electrocatalysts. MONOLITHOS hydrometallurgical leaching process was followed to recover Pt and Ir from the collected electrocatalyst powder (Figure 15). The used leaching system is based on inorganic solvents with low acidity, namely 3 M HCl, 4.5 M NaCl, $1\% v/v H_2O_2$. The use of NaCl (additional sources of ion chloride) helps to avoid the high acidity.





Figure 15. MONOLITHOS hydrometallurgical leaching process to recover CRMs from EoL electrocatalyst powder.

According to XRF analysis a leaching efficiency higher than 99% has been achieved for Pt and about 60% for Ir. Several experimental parameters are explored (HCI acidity, oxidizing agent concentration, solid/liquid ratio and reaction time) to achieve the highest as possible recovery yield of Ir. Leaching rate of Ir obtained using different experimental conditions (solid/liquid=50%, 4-6 M HCI, 1-3% v/v H_2O_2) will be determined and validated by ICP analysis in MONOLITHOS' facilities during next weeks.

Furthermore, a preliminary study of the delamination and separation of the catalyst layers from the EoL MEAs in an alcohol/water mixture has also been performed. More specifically, anodic and cathodic catalyst layers are separately subjected to obtain two different batches (Pt and Ir) of the PGMs (Figure 16). Firstly, the anode side of the MEA is wetted with solvents' solution and anodic electrocatalyst was separated and collected. Afterwards, the cathode side of the MEA was treated with a fresh solvents' solution and cathodic electrocatalyst was detached from the membrane and collected. Most of the electrocatalyst powders are detached from the membrane, while the process is under optimization and PGMs recovery is under investigation.





One side of the membrane immersed into the mixture → Nafion deformation



Second side of the membrane immersed into the mixture \rightarrow Nafion deformation

Figure 16. Studying the delamination and separation of the catalyst layers from the MEA to obtain two different batches (Pt and Ir) of the PGMs. In addition, during the last months MONOLITHOS has been granted with the patent with the title of "Process of recovery of critical metals from electrochemical stack devices" from Hellenic Industrial Property Organization with the application number: EL 245-0004386313. The same patent has been



submitted in the European Patent Office (1st of November 2022, Submission number: 11448195, Application number: EP22204905.8) waiting for its publication/assessment.