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Validation and characterization of suitable materials for bipolar plates in PEM water electrolysis

Manuel Langemann^{a,*}, David L. Fritz^a, Martin Müller^a, Detlef Stolten^{a,b}

^a Institute of Energy and Climate Research (IEK-3), Forschungszentrum Jülich, Germany

^b Chair for Fuel Cells, RWTH Aachen University, Germany

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ABSTRACT

The polymer electrolyte membrane (PEM) electrolysis cell is a promising prospect for the production of clean hydrogen by energy of renewable wind and solar sources. One component of the PEM electrolyzer is the bipolar plate (BPP), which serves as a multi-function component during PEM water electrolysis. Titanium is typically regarded as the state-of-the-art material. Mechanically it could potentially be replaced by lower-cost materials such as stainless steel, but under the harsh environmental conditions present in PEM water electrolysis, stainless steel is not corrosion-resistant and metal ions can dissolve. In this case metal ions would poison the catalyst and membrane, which leads to a reduction in the cell performance [1]. We have tested several coatings such as Au and TiN in PEM water electrolysis environments of varying severity for the application as a protective layer of bipolar plates. In order to determine possible candidates for a long-term test under real simulated PEM water electrolysis conditions, an experiment to determine pH value in PEM water electrolysis operation was developed to obtain the required pH value for the ex-situ testing of various coating materials.

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Introduction

The demand for renewable, green and sustainable energy has been increasing exponentially over the past decade [2]. To increase efficiency of installed renewable resources such as wind and solar energy, the off-peak energy that surpasses the grid demand must be captured. These renewable resources however tend to be quite intermittent and unpredictable, meaning that a large capacity, responsive and efficient storage system needs to be in place in order to fully utilize the installed renewable resources.

Polymer electrolyte membrane water electrolysis offers a good part-load performance to handle the fluctuations of the intermittent energy sources it will likely be coupled with. To maximize capital investment the idea of operating for short periods of time in an overload regime can drastically reduce costs. This does however come at a cost of operating at higher voltages and thus more corrosive environments.

To avoid the corrosion problems current cell design involves coating the titanium components with gold and platinum. This helps to avoid increased contact resistance from oxide layers that form on the titanium and from the hydrogen

* Corresponding author. Wilhelm-Johnen-Straße, 52428 Jülich, Germany. Tel.: +49 2461 61 9759; fax: +49 2461 61 6695.

E-mail address: m.langemann@fz-juelich.de (M. Langemann).

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embrittlement. These rare metal coatings on already expensive base metals drastically increase system costs to a point where the improved performance no longer outweighs the investment [3].

This work evaluates the alternative, less expensive metallic materials for the bipolar plate (BPP), which are to be used in the polymer electrolyte membrane water electrolyzer. The function of the BPP in the PEM cell is to ensure the media transport and the transfer of electrical current through the stack. Currently BPPs are made of titanium, this is however cost prohibitive and lower cost materials such as stainless steel e.g. AISI 304 or 316L could drastically reduce the overall system cost. Since stainless steel has a lower corrosion resistance in the PEM water electrolysis environment than titanium, the stainless steel substrate needs to be protected by a suitable coating against corrosion, which shows up as emission of metal ions absorbed by the membrane and leads to a decrease of cell performance [1]. The corrosion in the PEM water electrolysis cell is promoted by several factors, such as the applied potential, electrolyte parameters and the presence of pure oxygen. The cell operates with a Nafion® membrane in a temperature range of typically 50–90 °C. The minimum potential required for the decomposition of water at room temperature is 1.48 V (thermo-neutral voltage). Depending on factors like the quality of catalysts or the thickness and type of membrane, the required potential during operation could be raised to over 2.00 V. Additionally, the electrolyte properties, such as a low pH value, lead to an increased development of corrosion, which is expressed in a higher corrosion rate. The increased corrosion rates in the lower pH environments has been noticed in the fuel cell community despite the significantly lower potentials than those seen by an electrolyzer [4]. The demanding requirements of this particularly aggressive environment for the metallic BPP present great challenges in finding suitable materials at a relatively low cost.

The PEM electrolysis literature offers few hints to what the pH value of the circulating water in an operating cell or stack may be. Carmo et al. [2] mention the pH value within the cell can reach between 2 and 4, however this is referring to the catalyst layer and this is in direct contact/combined with the Nafion ionomer. Nafion is a fluorinated polymer with sulfonic acid side chains creating a very strong acid in the catalyst layer. This ionomer is however not in contact with the BPP and its pH value is therefore not an accurate representation of the environment visible to the BPP.

The fuel cell literature was also examined due to the similarities in many of the components between the PEM fuel cell and the PEM electrolyzer. This resulted in a large range of possible pH values from 1.0 to 0.001 molL⁻¹ H₂SO₄ used in corrosion testing [4]. The majority of the researchers have used a 1.0 molL⁻¹ H₂SO₄ electrolyte (sometimes with a few ppm F⁻) but it not specified how or why 1.0 molL⁻¹ H₂SO₄ was chosen [5–11]. The fluctuation in the pH value throughout the fuel cell literature, and the different operating conditions the two cells see does not clearly identify an appropriate electrolyte for the classification of coated metallic BPP for PEM water electrolysis.

In the literature, and by a series of electrochemical studies on AISI 304, it has been found that the pH value of the electrolyte has a critical influence on the degradation of the BPP material, which is shown by the corrosion rate [12]. Therefore

the knowledge of the suitable pH value is a requirement for the testing of any ex-situ corrosion in PEM water electrolysis and its determination is the primary focus of this work.

Experimental and procedures

A two-part experiment was developed to quantifiably classify possible coating materials for use in a PEM water electrolyzer. The first part is then designed to determine at what pH value this medium is in actual operation. The second part of the experiment is to determine the overall influence the pH value of the corrosive medium has on the corrosion rates at typical electrolysis potentials.

Determination of the pH value development of a running PEM water electrolysis cell

For the determination of the pH value, the cell was equipped with corrosion resistant bipolar plates to ensure that no corrosion reaction of the bipolar plate material influences the pH value measurement. The anode bipolar plate and the cathode bipolar plate were coated with Pt and Pt–Au, respectively. The extra Au coat added to the cathode bipolar plate was necessary to avoid Ti embrittlement by the produced hydrogen. On the anode side sintered titanium powder (T3P from GKN) was used as porous transport layer, and carbon paper (TPGH120 from Toray) was used on the cathode side. The homemade membrane electrode assembly (MEA) was fabricated using the blade-coat/decal method and contained 0.8 mg Pt₉₁₀₀ on the cathode and 2.2 mg Ir in the form of IrO₂ on the anode side. Nafion® N117 was used as membrane material. The hot-pressing step was conducted at 130 °C and 0.5 kN cm⁻². The MEA was afterwards conditioned in 0.5 molL⁻¹ sulfuric acid at 80 °C for 1 h and washed with fresh water till neutral pH value.

To determine the effective pH value of an operating PEM water electrolysis cell it is necessary to measure the pH value on both cell sides, on the oxygen electrode side (anode) and the hydrogen electrode side (cathode). Fig. 1 shows a realistic representation of the experimental setup of the pH value measurement.

Each electrode side was supplied by its own water tank, which consisted of a jacket glass bottle to ensure a specified temperature of the circulating water. PTFE was used as inert material for all tube connections. The unreacted water flowed back to the tank, where the pH value of the water was continuously measured. The signal of the InPro3250i/SG/225 pH electrode was recorded by a Mettler Toledo transmitter. The process data from the transmitter was logged using the Mettler Toledo software during the experiment. The pH electrodes are completely immersed in the water solution of both sides for the duration of the experiment. On both electrodes the volumetric flow rate of the cell feeding water was set as 15 ml min⁻¹. Before the experiment was started, the water was heated up to the operating temperature of 50 °C, which was provided by a Haake DC 30 thermostat.

The energy for the cell reaction is delivered by a power supply (TDK-Lambda GEN 20-76). For the pH measurement a constant current density of 0.5 A cm⁻² was set. This value

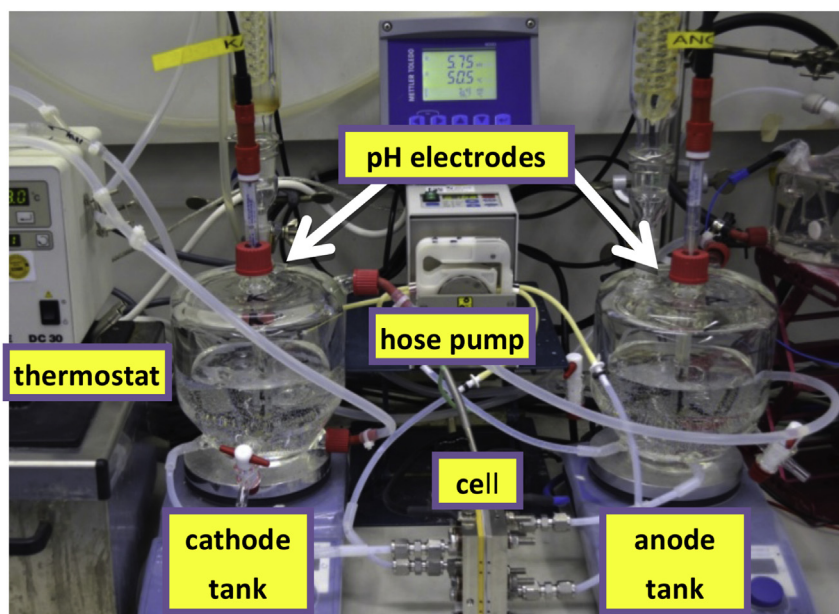


Fig. 1 – Realistic demonstration of the pH value measurement setup.

corresponds to a voltage of 1.67 V in the beginning and 1.71 V at the end of the measurement. The water tanks are each equipped with a reflux condenser to minimize volume losses through the evaporation of the reaction water. At the beginning of the experiments the fresh water exhibits a pH value of ca. 5.6. The experiment was performed for 50 h. In Table 1 the pH values of the measurements are listed.

Experimental setup of the electrochemical characterization methods and electrochemical measurement procedure

The electrochemical measurements were arranged in a corrosion measurement cell, which was designed in the Forschungszentrum Jülich at the IEK-3 (Fig. 2). The samples were measured in a three-electrode set-up inside the glass cell. The working electrode (WE, metallic sample) was pressed by a stamp against the PTFE wall and is sealed off by a Viton gasket. The PTFE wall exhibits a circular opening for an active WE area of 15.21 cm², which is in contact with the electrolyte. A Platinum mesh was used as counter electrode material and placed near to the WE. As reference electrode a HydroFlex[®] reversible hydrogen electrode (RHE) from Gaskatel (Germany) was used.

The samples were ultrasonically cleaned in an acetone bath for 30 min prior to the electrochemical measurement. After the cleaning process the samples were connected to the Autolab potentiostat PGSTAT302N (Metrohm). The electrolyte, which was used for the initial testing consisted of 0.5 molL⁻¹ sulfuric acid at a temperature of 50 °C. In the beginning of the measurement the samples were electrochemically cleaned up

in a cyclic voltammetry process by 10 cycles between a potential range from –0.5 ... 1.5 V (vs. RHE), whereby in the end the samples were hold for 30 s at a potential of –0.5 V (vs. RHE) to remove adsorbed oxygen.

After the cleaning process the LSV procedure was directly started and swept through a potential range of –0.5 ... 2.0 V (vs. RHE) with a scan rate of 0.005 V s⁻¹. A transformation of the received data in a half logarithmic plot delivers the intersection point by extrapolating the Tafel lines. The intersection point represents the corrosion current at open circuit voltage. Following the LSV procedure, the CA procedure was started at a constant potential of 2.00 V (vs. RHE) during 3 h or

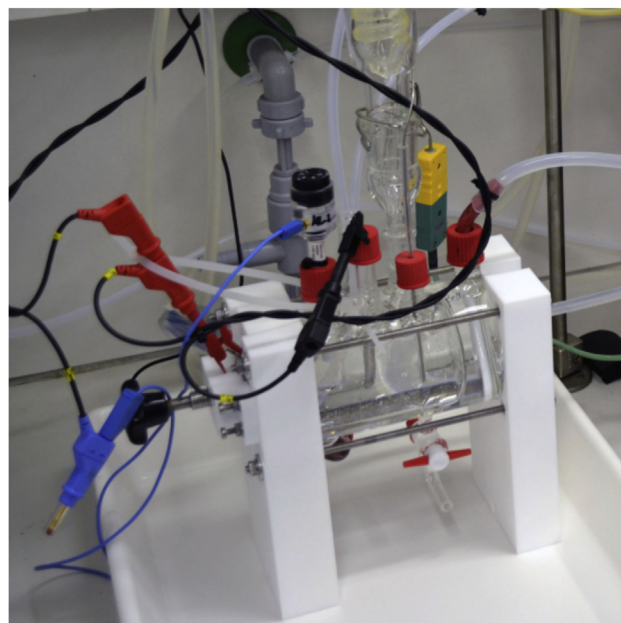


Fig. 2 – Setup of three-electrode measurement to characterize metallic samples.

Table 1 – Lists of pH value measurement.

Procedure	Time [h]	pH _{an} (begin)	pH _{an} (end)	pH _{ca} (begin)	pH _{ca} (end)
1	50	5.60	3.49	5.55	4.54
2	50	5.50	3.44	5.54	4.57

24 h. After the initial proof-of-concept for the corrosion test cell a series of tests were performed each at an increased pH value to understand how sensitive the corrosion rates are to the pH value of the electrolyte. This required longer CA measurement times for the higher pH value experiments. The times and pH values for the range of electrolytes tested can be seen in Table 2.

In order to evaluate the individual coatings qualitatively the coated substrates must be evaluated in the ex-situ corrosion test cell. This requires an in-depth knowledge of the pH value of an operating electrolysis system to avoid testing the incorrect corrosive mechanism. Thus an in-situ pH value measurement was designed to obtain the realistic operating conditions. The following section details the steps taken to determine the pH value of the circulating water in an operating PEM electrolysis system.

Results and discussion

The results of the measurement of the pH value development and the material characterization are presented herein. Along with the material characterization the influence of the electrolyte pH on the corrosion rate is also studied.

Evaluation of the pH value development

In Fig. 3A the development of the pH value in the feeding water of a working PEM water electrolysis cell is presented over time.

To measure the pH value on both sides 1000 ml of MilliQ water was added to the anode tank and 250 ml was added to the cathode tank. The difference in starting volumes is due to losses of the water consumption during the electrolysis reaction and water transport across the membrane via electro-osmotic drag across the Nafion membrane. This transport rate can be calculated and is found to be between 3 and 4 water molecules transferred from the anode to the cathode per diffused proton [13–15]. In our experiment the average water transfer rate was 12.53 ml h^{-1} (1st run) and 13.74 ml h^{-1} (2nd run). The pH value of the first measurement procedure starts at ~5.6 on both sides and it drops during operation. In the initial phase of experiment (first 5 h at Fig. 3A) the pH values on both sides first fall and then rise at different time scales, due to the stabilization of the pH sensors to the new medium. It is assumed, that the measurement of proton concentration needs time to equilibrate and this occurs more quickly on the cathode side due to its initial lower water volume. After reaching its maximum, the pH values drop slowly

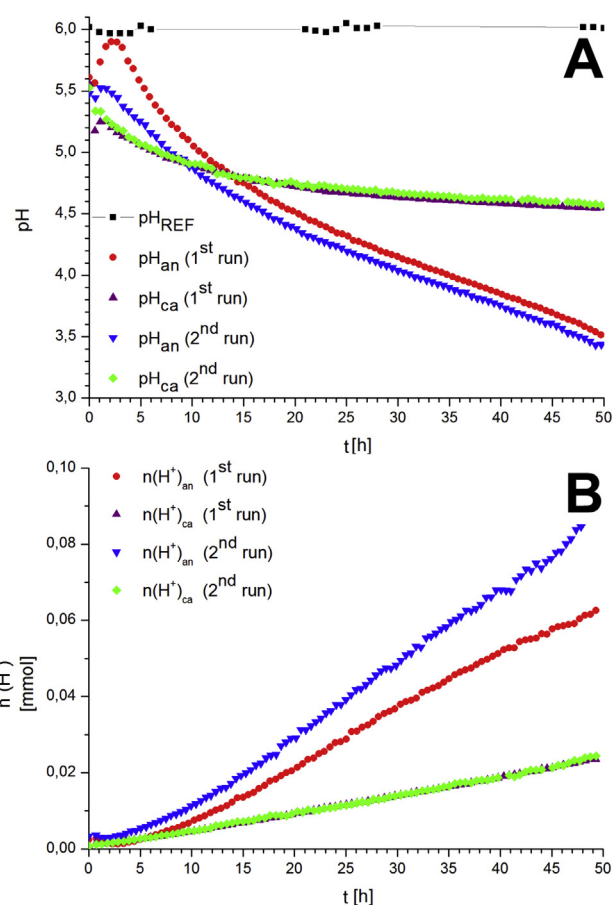


Fig. 3 – (A) Development of the pH value in the feeding water of a PEM water electrolysis single cell. The pH values were obtained for the anode and cathode side independently. (B) Development of the proton entry in the water (normalized by water volume).

on both sides. The initial and final volumes on each side of the cell are presented in Table 3.

It is noticeable in Fig. 3A that the anode side has a steeper reduction in pH over time than the cathode side. This could mean a higher proton entry rate on the anode side. However, since the water drag is constantly reducing the volume of water on the anode side, it is also possible that the difference in rates is caused by the constantly changing volume. At the end of the 50 h run, the pH value of the anode side reached a value of 3.5 and the cathode a value of near 4.5. Fig. 3B illustrates the development of the normalized proton content for cathode and anode side. It is clear to notice that the normalized proton content of the anode side is greater than that of the cathode side.

Table 2 – List of the measurements in corrosion cell.

Substrate	Coating	Time [h]	Potential [V]	pH
AISI 304	TiN	3	2.00	0.48
AISI 304	Au	3	2.00	0.48
AISI 304	TiN	24	2.00	2.89
AISI 304	Au	24	2.00	2.89

Table 3 – Volume of water of both electrode sides.

Procedure	V _{an, begin} [in ml]	V _{an, end} [in ml]	V _{ca, begin} [in ml]	V _{ca, end} [in ml]
1	1000	204	250	856
2	1000	187	250	913

Additionally there is an increase in proton content in the second run relatively to the first procedure. For the MEA fabrication, the Nafion membrane is conditioned in sulfuric acid to protonate the sulfonic groups that are previously obtained in the Na⁺ form. Additionally, 15–30% of Nafion ionomer can be found in the catalysts layer, on both sides of the MEA. The water that is fed into the cell can potentially wash out traces of sulfuric acid and ionomer from the MEA, potentially increasing the pH of the feeding water, especially in the first hours of cell operation.

For the second procedure the water from the initial experiment was completely removed from the cell and the water tanks. Fresh MilliQ water was then refilled. The second procedure of measurement also ran for 50 h. During this time the development of pH value of the anode side follows a similar trend to the anode side of the first procedure. There is only a pH value offset of ~ 0.2 for the entirety of the second procedure. A change in pH value caused by a formation of carbonic acid could be excluded, because a pH measurement of a reference tank, which was heated to 50 °C show no changes in the pH value during an operating time of 50 h.

The time frame of 50 h was chosen to demonstrate the development of the changing pH value and how the pH value behaves in the initial phase of operation for the PEM water electrolysis cell. The 50 h time was chosen arbitrarily, and unfortunately did not lead to a constant pH value level in the end of measurement. We can easily assume, that the pH value will continue to drop. However, it is not clear at this stage how long it will take to stabilize. The results of these tests are important for designing the operation strategy and system supply in PEM water electrolyzers. Operating with only fresh water and not recirculating is too costly, while only using the minimum amount of water and only recirculating will result in increased corrosion rates. By understanding the rate at which the pH value changes, an initial volume and a fresh water introduction rate can be selected for the system to reduce degradation rates. Because the controlling mechanism of the changing pH value is suspected to be related to the washing out of different compounds found in the MEA, the operational strategy, namely the addition of fresh water to the system will be much larger during the initial break-in period of a cell. The determination of an exact break-in period is dependent on many variables and operational parameters and beyond the scope of this article; however through the ongoing research such a correlation looks promising.

Polarization curves

The characteristics of the IV-curves for the AISI 304 stainless steel samples (same batch) in different electrolyte solutions with varied concentration of sulfuric acid is shown in Fig. 4.

For the material characterization of bipolar plates used in PEM water electrolyzers the examined cell voltage range between 1.6 and 2.0 V is crucial. Since the water splitting reaction will start at 1.23 V, and decent performances can generally be obtained with cell voltages higher than 1.6 V, 1.6 V becomes the threshold. The curve development of 0.5 molL⁻¹ H₂SO₄, which is compared to the curve of 0.001 molL⁻¹ H₂SO₄ at 1.7 V is over 80 times higher in current density. This value is however, not only allocated to the

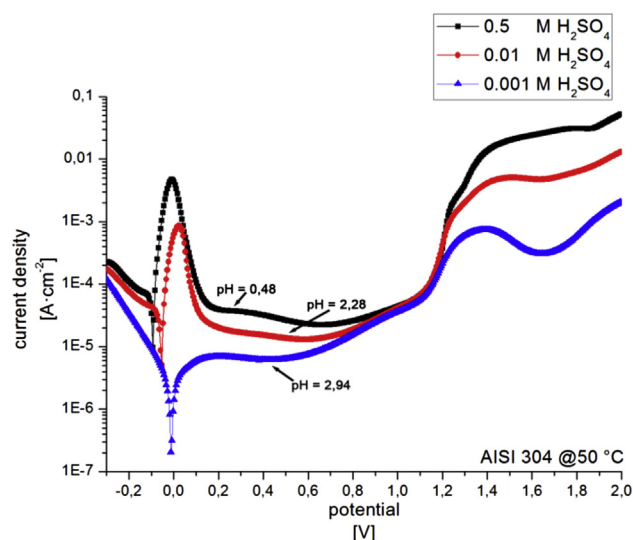


Fig. 4 – Polarization curves for AISI 304 with varied pH values at 50 °C.

corrosion current, it also contains the current density of the oxidation evolution reaction (OER), which starts above 1.2 V. It is clear that the curve development is influenced by electrolyte parameters such as the pH value, because all the other parameters are kept constant, e.g. temperature, potential area and scan rate. In this case it means the smaller pH value the higher the corrosion current inside the active corrosion area (peak in the curve at 0 V). The different corrosion currents are listed in Table 4.

When comparing the current densities of the 0.5 molL⁻¹ and 0.001 molL⁻¹ sulfuric acid solution it is obvious that the corrosion development of AISI 304 is almost 20 times higher in 0.5 molL⁻¹ than in the 0.001 molL⁻¹ sulfuric acid solution.

It is therefore essential to know the exact proton concentration of the applied electrolyte. For this reason it was necessary to in-situ evaluate the exact development of the pH value in a PEM water electrolysis cell. This knowledge also aids in the selection of a corrosive media for accelerated testing while avoiding different corrosive mechanisms that could present erroneous results.

Degradation test by a constant potential

To evaluate Au or TiN coated stainless steel bipolar plates, the CA procedure was started at a constant potential of 2.00 V in 0.5 molL⁻¹ sulfuric acid during 3 and 24 h. The summarized data can be seen in Table 2. Fig. 5 shows the bipolar plates used in the corrosion tests. The used coatings are Au and TiN, which were coated by PVD technique with a deposit thickness

Table 4 – Display of the corrosion current densities.

Electrolyte H ₂ SO ₄ [mol L ⁻¹]	pH	Corrosion current density[μA cm ⁻²]
0.5	0.48	54.5
0.01	2.28	32.3
0.001	2.94	2.8

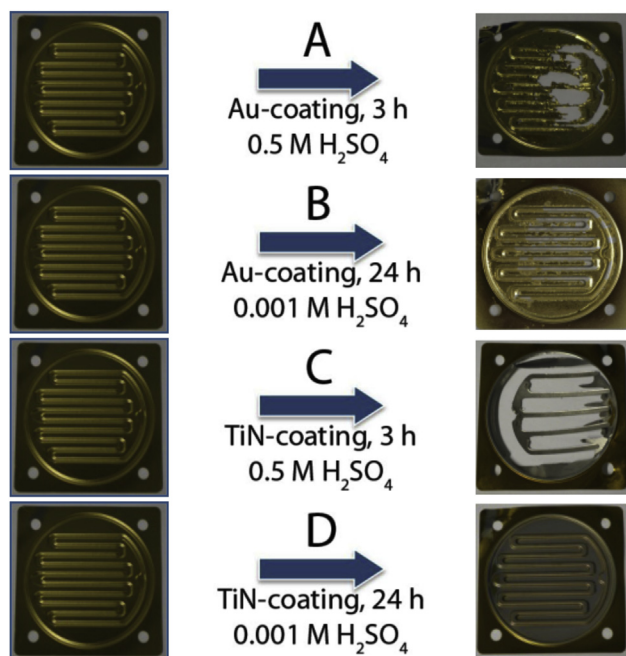


Fig. 5 – Development of Au and TiN coated AISI 304 sample shapes in 0.001 M and 0.5 M sulfuric acid after 3 h or 24 h treatment.

of 0.2 (Au) and 0.5 (TiN) μm . Fig. 6 shows the corrosion development in the different samples after their exposure to the corrosion media. As shown in Fig. 5 A and C in high acidity solutions, where the samples underlie a stronger corrosion development than the other samples in B and D tested in a milder corrosion solution.

The exposure to the milder corrosion media ($0.001 \text{ molL}^{-1} \text{ H}_2\text{SO}_4$) is responsible for attacking only the coated layers, and the substrate materials remain stable throughout the duration of the test.

In the initial surface analysis of untreated Au and TiN coated samples by SEM it was determined, that small areas on the surface were not fully coated, resulting in pin-holes in the

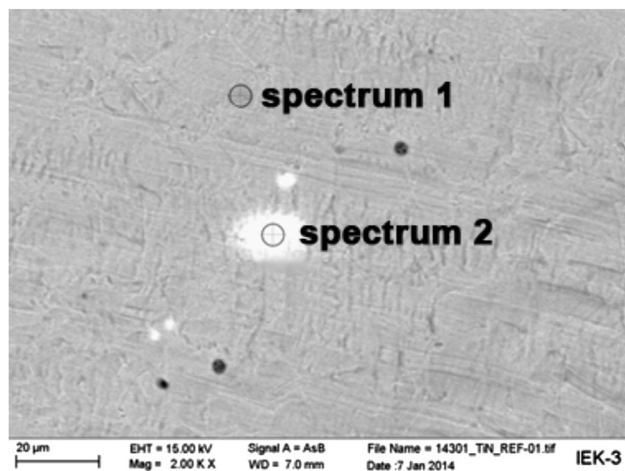


Fig. 6 – SEM micrograph of an untreated SS 304 coated with TiN.

Table 5 – EDX data (in wt-%) of a SS 304 sample coated with TiN.

Element	Spectrum 1	Spectrum 2
Ti	25.9	1.54
N	17.7	—
Si	—	0.49
Fe	40.5	63.9
Cr	11.6	17.4
Ni	4.3	8.5

coating. The SEM micrograph of TiN coated SS 304 is visible in Fig. 6 and the corresponding values of the EDX spectra 1 and 2 are shown in Table 5.

Looking at the values in Table 5 it is clear that at the position of spectrum 2 very little Ti and no N could be determined. This is an indication of defective spots in the TiN layer.

The corrosion media undercut these pits and directly attack the substrate material. Due to the instability of stainless steels to high acid concentrations the substrate material as a result is dissolved by the corrosion media.

Conclusion

In the first part of this work it is shown that the pH value of the recirculated water plays a vital role in the stability of the metallic components on the anode side of the PEM water electrolysis cell. It was then found that during normal cell operation the pH value drops significantly creating an increasingly more acidic environment that risks the stability of the metallic components already under high potential ($>1.48 \text{ V}$) on the anode side.

The effect of the increased acidity ($\text{pH} = 5.6$ to $\text{pH} = 3.4$) was determined through the polarization curves of various acidic media where the corrosion current was shown to depend greatly on the pH value. Under the high potential of the PEM water electrolysis operation ($\sim 2.00 \text{ V}$) the pH value is the dominant factor influencing the long term stability of the substrate materials and their coatings.

The development of the pH value gives great insight on the main cause of the corrosion problems in PEM water electrolysis. By understanding the mechanisms causing this acidic environment, not only can better accelerated aging tests be developed, but steps can be taken to reduce the overall corrosion rates developing operating strategies in a new system.

As an outlook, we will expand our measurements to determine the cause for the proton entry in the water. A possible method could be the determination of the pH value development in a cell operation without current (only circulating water). Another step is maintaining a mild pH value by extrapolating of the pH value change over time and adding fresh water to keep proton concentrations constant.

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