Composition of highly concentrated silicate electrolytes and ultrasound influencing the plasma electrolytic oxidation of magnesium

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Abstract. Magnesium and its alloys are increasingly in use as lightweight construction materials. However, their inappropriate corrosion and wear resistance often prevent their direct practical use. The plasma electrolytic oxidation (PEO) is a promising, environmentally friendly method to improve the surface characteristics of magnesium materials by the formation of oxide coatings. These PEO layers contain components of the applied electrolyte and can be shifted in their composition by increasing the concentration of the electrolyte constituents. Therefore, in contrast to the use of conventional low concentrated electrolytes, the process results in more stable protective coatings, in which electrolyte species are the dominating constituents. In the present work, the influence of the composition of highly concentrated alkaline silicate electrolytes with additives of phosphate and glycerol on the quality of PEO layers on the magnesium alloy AZ31 was examined. The effect of ultrasound coupled into the electrolyte bath was also considered. The process was monitored by recording the electrical process variables with a transient recorder and by observation of the discharge phenomena on the sample surface with a camera. The study was conducted on the basis of a design of experiments. The effects of the process parameter variation are considered with regard to the coatings thickness, hardness and corrosion resistance. Information about the statistical significance of the effects of the parameters on the considered properties is obtained by an analysis of variance (ANOVA).

1. Introduction
Magnesium alloys are increasingly in use as lightweight construction materials. Their low density, high specific strength and good damping behaviour against mechanic vibrations and electromagnetic oscillations make these alloys interesting for applications in automotive and aerospace industries as well as housing materials for high-quality electronics. However, the insufficient resistance against corrosion and wear often prevent the direct practical use of magnesium materials.[1–2] The plasma electrolytic oxidation is a promising environmentally friendly surface treatment method to handle these challenges. During the process, the workpiece is immersed in an aqueous electrolyte, opposed to a cathode and receives an increasing anodic polarisation. This procedure leads to the formation of an anodic film on the material surface and to the development of a gas-steam shell enveloping the workpiece geometry. The voltage drop over the cell arrangement is concentrated between the metal substrate and the gas-steam shell/electrolyte interface. Afterwards, this equipotential area called quasi
cathode becomes the starting point of spark discharges from the electrolyte through the gaseous phase and the anodic film into the substrate. The discharges leave oxidized spots and produce protective oxide ceramic PEO layers by further oxide formation, melting and remelting processes subsequently. [3] This method is well understood and commercially used for aluminium materials. The excellent technical characteristics of the developing alumina allow for the formation of hard and resistive coatings. In contrast, the PEO layers on magnesium materials consist mainly of magnesia, which has less protective properties. In addition to oxide compounds of the substrate material, the PEO layer contains varying amounts of electrolyte constitutes in dependency of its composition and concentration. For the plasma electrolytic oxidation of magnesium materials, substances with the anions \( \text{AlO}_2^- \), \( \text{SiO}_3^{2-} \), \( \text{PO}_4^{3-} \), \( \text{F}^- \) and \( \text{SnO}_3^- \) are common electrolyte components. Thus, the formed oxide ceramic coatings can contain significant amounts of Al, Si, P, F and Sn [4–7].

Ultrasound is a common process parameter to improve the results of wet-chemical surface treatment methods. On the field of the anodic surface modification, a few publications describe the positive influence of ultrasound on the process characteristics and treatment results. According to these studies, ultrasound coupled into the electrolyte bath during the conventional anodic oxidation can increase the growth rate of anodized layers on aluminium in oxalic acid [8] or support the formation of a regular cell structure while anodizing titanium in nitric acid [9]. For plasma electrolytic oxidation of aluminium it has been reported that ultrasound can increase the compactness and the \( \alpha-\text{Al}_2\text{O}_3 \) amount of the PEO layers [10] and decreases the formation voltage during the process [11]. Concerning the influence of ultrasound on the PEO process on magnesium, there are hardly any data, but first studies for the alloy ZK61 also indicate a positive influence on the resulting corrosion resistance [12].

In our present work the magnesium alloy AZ31 was used. The study follow the research approach of focussing on the incorporation of electrolyte constitutes into the PEO layer to shift its composition in favour of more stable phases than MgO. The main component of the electrolyte systems used is silicate in combination with the additives phosphate and glycerol. Figure 1 gives a graphical representation of the composition of common low concentrated electrolytes for the PEO process on magnesium, based on the mentioned constitutes (a) and in direct comparison to the electrolyte mixtures used in the present study (b).

![Figure 1](image_url)

**Figure 1.** Chemical compositions of common low concentrated electrolytes (a) and the high-concentrated electrolytes of this study (b) for the PEO treatment of magnesium.
To ensure the environmental compatibility the use of fluoride was renounced. To investigate a possible influence of ultrasound on the process, ultrasound is coupled into the electrolyte bath by pulse mode.

2. Experimental Methods

The magnesium alloy AZ31, the composition of which is given in table 1, was used as the sample material for all tests.

**Table 1. Chemical composition of the AZ31 alloy.**

| Material | Allying elements (wt%) |
|----------|------------------------|
|          | Mg  | Al  | Zn  | Ti  | V   | Fe  | Mn  |
| AZ31     | Balance | 2.5–3.5 | 0.6–1.4 | ≤ 0.05 | ≤ 0.005 | ≤ 0.005 | ≤ 0.2 |

The single samples had a cylindrical geometry with a diameter of $d_s = 20$ mm and a height of $h_s = 10$ mm, and were included into the electrical circuit by a magnesium wire with a diameter of $d_c = 3.2$ mm laterally pressed in (this electrical contact is visible in figure 7 on the right side of the AZ31 samples). Thus, the active sample surface results as $A_s = 1248.6$ mm². The magnesium wire by itself was insulated electrically against the electrolyte. The current-regulated electrical regime was applied by a rectifier (Plating Electronics, Germany) providing effectively 550 V and 50 A at its maximum. The current pattern was predefined as a symmetric pulse of $I_1 = I_2 = 2$ A with durations of $t_1 = t_2 = 5$ ms. The resulting current densities were $i_1 = i_2 = 16$ A/dm².

The electrolytes were prepared with deionised water, different amounts of Na₂SiO₃ · 5 H₂O, Na₂HPO₄, and C₃H₅(OH)₃. The pH value was between 13.1 and 13.5. Therefore, the phosphate ions were present as PO₄³⁻ rather than HPO₄²⁻. The ultrasound was coupled into the electrolyte bath by a sonotrode (Weber Ultrasonics, Germany) with frequency of 30 kHz and a power output of 78 W by pulse mode. The concentrations of the three electrolyte components and the duration of the ultrasound pulse were varied on three different levels with a logarithmically oriented graduation. A design of experiments (DOE) according to the Box-Behnken design [19] was used. Table 2 gives an overview of the parameters, their notations, and level values. Figure 2 shows a graphical representation of the experimental plan.

**Table 2. Overview of the varied process parameters.**

| Parameter               | Notation | Levels 1 | Levels 2 | Levels 3 |
|-------------------------|----------|----------|----------|----------|
| Ultrasound pulse duration (s·min⁻¹) | U        | 1        | 8        | 60       |
| Silicate concentration (mol·l⁻¹) | S        | 1.00     | 1.32     | 1.75     |
| Phosphate concentration (mol·l⁻¹) | P        | 0        | 0.035    | 0.09     |
| Glycerol concentration (mol·l⁻¹) | G        | 0        | 0.02     | 0.04     |
In the further article the capital letters U, S, P and G in combination with the corresponding parameter level are used as short nomenclature to describe the parameter set of a certain experiment. For example the parameter set of experiment 1 with an ultrasound pulse of 1 s, followed by pulse pause of 59 s and an electrolyte composition of 1.00 mol/l silicate, 0.035 mol/l phosphate, and 0.02 mol/l glycerol is abbreviated as U=1, S=1, P=2 and G=2.

To enable a statistical support of the obtained results, every experiment was carried out three times. Furthermore, a randomisation of the experiment order is desirable to minimize the influence of experimentally not determinable disturbances. Therefore and to limit the experimental effort, the electrolytes were prepared by systematic up-concentration. Within a certain electrolyte the experiments with different ultrasound pulse modes were carried out in randomised order. The electrolytes were prepared with a volume of 5 liters and filled in a cuboidal acrylic glass container, which was surrounded by cooling water with a temperature of 5 °C. All chemicals used: sodium metasilicate pentahydrate (Sigma-Aldrich, Germany), disodium phosphate (Merck, Germany) and glycerol (Roth, Germany) were of an analytical grade.

To observe the development of the electrical process variables, a transient recorder DL850 (Yokogawa, Japan) was connected to the electrical circuit. The voltage signal was acquired directly from the cell electrodes. The current flow was calculated based on Ohm’s law and the recorded voltage drop over a measuring resistor of $R_m = 1 \Omega$, which was included in the cathode strand. The electric measurements had a measuring interval of 1 min, a measuring duration of 200 ms and a sampling rate of 100 kS/s. The measured values of the electrical process observation were processed by a low-pass filter with a cut-off frequency of 15 kHz. The resulting data give a statement about the development of the maximum anodic and cathodic voltage during the processes. In addition, they enable to check whether the predefined current form has been correctly transferred to the experimental setup by the rectifier. To monitor the discharge behaviour on the sample, a camera GoPro Hero 3 (GoPro, USA), was immersed into the cooling water and oriented to the sample surface. Figure 3 shows a schematic representation of the complete experimental setup.
Figure 3. Schematic representation of the experimental setup.

The evaluation of the recorded videos allows determining to which extent the variation of the investigated process parameters influences the occurrence of certain discharge stages as well as their temporal transition to one another.

After the PEO treatment, the three samples of every experiment were cut into two halves. Two different samples of every experiment were prepared to cross-sections. Afterwards, these cross-sections were used for measurements of the layers thickness and Martens hardness at an optical microscope and a hardness measuring instrument Fischerscope HM2000 XYm (Fischer, Germany). The layer thickness $d$ was determined by 10 measurements at each cross-sections. The Martens hardness $HM$ was measured to examine a possible influence of the process parameter variation on the mechanical characteristics of the PEO-layer material. The measure force of 50 mN was applied in 30 s and kept constant for further 30 s. A Vickers diamond was used as indenter. The measurements were conducted 5 times at each cross-sections. To minimize a falsifying influence of the PEO-layers’ morphology, the indents were placed in compact regions of the coatings. To investigate the corrosion behaviour of the PEO layers, current density potential measurements were carried out for each repetition of the single experiments. The curves were recorded in a three-electrode arrangement, in which the test sample is the working electrode with an active surface area of 0.785 cm$^2$. A platinum sheet was used as counter electrode and a Ag/AgCl electrode as reference electrode. The used test solution was 0.1 mol/l NaCl, had a volume of 250 ml and a temperature of 25 °C. The potential was varied from -1800 mV up to -600 mV with a potential feed of 1 mV/s. Afterwards, the Butler-Volmer equation, which is shown in equation 1, was fitted manually with a MATLAB tool on the measured data by varying $i_0$ and $\alpha$.

$$i(i_0, \alpha) = i_0 \cdot \left[ \exp \left( \frac{\alpha \cdot z \cdot F \cdot (\varphi - \varphi_0)}{R \cdot T} \right) - \exp \left( - \frac{(1 - \alpha) \cdot z \cdot F \cdot (\varphi - \varphi_0)}{R \cdot T} \right) \right]$$
In this equation, $F$ and $R$ are the Faraday and the universal gas constant, respectively. $T$ is the absolute measuring temperature and $z = 2$ is the charge number according to the oxidation state of magnesium ions. The quantity $a$ describes the transfer coefficient of the anodic and cathodic corrosion reaction. The corrosion current density $i_0$ quantifies the net current flow at the open circuit potential and can be obtained from the fit. It provides an integral and well comparable statement about the corrosion properties and defect densities of the produced coatings, and is used for the evaluation of the experiments in this study. Figure 4 shows a diagram with a measured and fitted current-density potential curve. The dashed lines show the corresponding Tafel plots, which intersection represents the corrosion current density.

![Figure 4. Measured $i(U)$ curve and fitted $i(U)$ curve according to the Butler-Volmer equation.](image)

Information about the statistical significance of the effects of the parameter variations on the maintained layer properties are generated by an ANOVA. Further investigations respecting the layers morphology, chemical composition and phase structure were conducted at selected samples. The layers morphology was examined at a scanning electron microscope (SEM) 1455VP (LEO, Germany) using an excitation voltage of 25 kV based on back-scattered electron (BSE) images, which show an element specific contrast. The chemical compositions of the investigated coatings were measured in dependency of the substrate distance $d_{\text{substr}}$ by an energy-dispersive X-ray spectroscopy (EDX) system included in the scanning electron microscope. X-ray diffraction (XRD) measurements were carried out to determine the coating’s phase structure. Therefore, an X-ray diffractometer D8 Discover (Bruker AXS, USA) was used. The samples were measured with Co-$K_{\alpha}$ radiation in a line focus with Bragg-Brentano geometry and a Lynxeye-XE detector (Bruker AXS, USA) on a length of 10 mm for 3.5 h. The phase allocation was based on the PDF-2 data base of the International Centre for diffraction Data (ICDD).
3. Results and Discussion

Figure 5 shows representative current density and voltage curves of the applied bipolar pulse regime. The black dashed line describes the predefined current pattern, the solid line displays the measured curve. The grey graph indicates the corresponding measured voltage signal.

![Figure 5](image)

**Figure 5.** Predefined current-density curve (dashed black line) and representative measured current-density (solid black line) and voltage (dashed black line) curves of the applied bipolar pulse regime.

The measured current-density course shows a significant deviation from its predefined pattern. This can be explained by the fact that the absolute current values of these processes are at the lowest part of the working range of the used rectifier, where its working accuracy is restricted. As a result, the applied current pattern in this study was a kind of transition form between triangular and rectangular pulse, which has to be taken into account for reproduction or upscaling of these processes. Figure 6 shows envelopes of the course of the maximum anodic and cathodic voltages of the three repetitions of the experiment couples 9/10 (a), 15/16 (b), 7/8 (c), and 9/11 (d). At these experiment couples, three experimental parameters were kept constant whereas one was shifted from level one up to level three (see graphical representation of the DOE at figure 2).

The influence of the parameter variations on the resulting process voltages is of interest for economic considerations respecting the total consumption of electrical energy. Obviously, the parameters ultrasound and phosphate concentration do not influence the voltage courses during the PEO process (figure 6 (a) and (c)). A higher silicate concentration lowers the anodic process voltage by approximately 50 V (figure 6 (b)). An elevated glycerol concentration (figure 6 (d)) increases the anodic process voltage by 20–50 V. It also prevents the appearance anodic voltage sags like shown in figure 6 (d) in the process time between 300 and 400 s. This observation can be made also for the experiment couples 6/8, 17/19, and 20/18, which also differ only with respect to the shift of the glycerol concentration from level one up to level three. This suggests that the electrolyte additive glycerol has a stabilizing effect on the process in addition to its anodic voltage increase. Whether there is indeed a positive influence on the resulting layer properties can be determined by the investigations below.
Figure 6. Envelopes showing the course of the maximum anodic and cathodic voltages over the three repetitions of the experiment couples 9/10 (b), 15/16 (b), 7/8 (c) and 9/11 (d); the experimental parameters of these couples were similar except for one process parameter, which was varied from level one up to three.

The evaluation of the recorded video data allows for a categorization of the discharge behaviour during the process into three different stages, which are depicted representatively for the example of experiment 23 in figure 7 (a–f). Stage I shows high-frequency, uniformly distributed, bright little discharges (figure 7 (a)–(b)). Stage II shows high-frequency, uniformly distributed, weakly luminous little discharges accompanied by rare low-frequency, single, bright big discharges (figure 7 (c)–(d)). Stage III shows low-frequency, localized, bright big discharges (figure 7 (e)–(f)).

Figure 7. Exemplary photographs with process time (mm:ss) for discharge stages I (a)–(b), II (c)–(d), and II (e)–(f), recorded from experiment 23, obtained by the optical process monitoring.

Figure 8 (a) shows an effect diagram, which represents the parameters influence on an investigated quantity, in this case the transition time of discharge stage I into discharge stage II $t_{I \rightarrow II}$. The bar plots show the effect of every single investigated process parameter. The error bar includes the process parameter variation over the complete experimental plan. The value of the error probability $p_{\text{error}}$ is obtained by the ANOVA and provides a statement whether the observed effect is statistically significant or not. In this study, a confidence interval of 5 % is chosen. Thus, parameters with a $p_{\text{error}}$ of
lower than 5% are considered as statistically significant. The ultrasound as well as the silicate and phosphate concentration accelerate the transition by raising the parameter level, while an increased glycerol concentration delays it. All effects are statistically significant. Figure 8 (c)–(d) shows effect diagrams for the influence of the investigated parameters on the layer properties thickness, martens hardness and corrosion current density.

![Figure 8](image)

**Figure 8.** Effect of the investigated parameters on the transition time between the discharge stage I and II (a) and the layer thickness (b), hardness (c) and corrosion current density (d).

A higher layer thickness is supported by increasing the silicate and glycerol concentration, while the phosphate concentration results in a maximum at level 2. The ultrasound coupling had no statistically significant effect (figure 8 (b)). The hardness of the PEO layers is increased by raising the glycerol concentration level, and shows maximum values for ultrasound pulses on level one and silicate concentrations on level three. The phosphate concentration has no statistically significant influence (figure 8 (c)). In comparison to the untreated substrate material, represented by the dashed line, the hardness clearly increases by the PEO treatment. Figure 8 (d) shows the obtained data for $i_0$ where a low value is desirable. The corrosion current density is minimized by increasing the phosphate and glycerol concentration. The silicate concentration leads to reduced values at level 1 and 3 while ultrasound has no statistically significant influence. In comparison to the untreated substrate material, represented by the dashed line, the corrosion current densities are clearly lowered by the PEO treatment. The PEO layer of the experiments 11, 12 and 16 show the lowest corrosion current densities, which indicates that they possess the lowest defect densities. Additionally, they show high values for layer thickness and hardness. Table 3 gives a quantitative overview of this optimal experimental parameters and the resulting layer properties. Figure 9 shows backscatter electron micrographs of the corresponding cross-sections.
Table 3. Quantitative overview of the optimal parameters obtained by this study and the resulting layer properties.

| Experiment | Parameters ultrasonic ultrasound (s·min⁻¹) | \(c_{\text{silicate}}\) | \(c_{\text{phosphate}}\) (mol·l⁻¹) | \(c_{\text{glycerol}}\) | \(i_0\) (10⁻⁴ mA·dm⁻²) | \(HM\) (N·mm⁻²) | \(d\) (µm) |
|------------|---------------------------------------------|----------------|---------------------------------|----------------|----------------|----------------|-------------|
| 11         | 1                                           | 1.32           | 0.035                           | 0.04           | 3.5 ± 1.3      | 3600 ± 200    | 43 ± 18     |
| 12         | 60                                          | 1.32           | 0.035                           | 0.04           | 3.1 ± 1.2      | 3700 ± 300    | 58 ± 21     |
| 16         | 8                                           | 1.75           | 0.090                           | 0.02           | 2.5 ± 0.2      | 3700 ± 200    | 64 ± 14     |

Figure 9. BSE micrographs of the substrate (S) and PEO layers (L) of the experiments 11 (U=1, S=2, P=1, G=3), 12 (U=3, S=2, P=2, G=3) and 16 (U=2, S=3, P=3, G=2).

The micrographs show that the PEO layers consist of an inner zone with a fine porosity and an outer zone with areas with a relative compact structure combined with big porosities. The results of EDX measurements with increasing substrate distance \(d_{\text{substr}}\) are depicted in table 4.
Table 4. Chemical composition in dependency of the distance from the substrate for the PEO layers of the experiments 11 (US=1, S=2, P=2, G=3), 12 (U=3, S=2, P=2, G=3) and 16 (U=2, S=3, P=3, G=2).

| Experiment 11 | Experiment 12 | Experiment 16 |
|---------------|---------------|---------------|
| $d_{\text{substr.}}$ (µm) | Element content (at%) | $d_{\text{substr.}}$ (µm) | Element content (at%) | $d_{\text{substr.}}$ (µm) | Element content (at%) |
| O  | Si  | Mg  | Na  | O  | Si  | Mg  | Na  | O  | Si  | Mg  | Na  |
| 5  | 56  | 16  | 25  | 2  | 12  | 6   | 18  | 17  | 3  | 4   | 58  | 15  | 23  | 3  |
| 24 | 60  | 21  | 13  | 4  | 35  | 61  | 21  | 15  | 3  | 20  | 60  | 22  | 14  | 4  |
| 46 | 58  | 19  | 16  | 6  | 45  | 61  | 20  | 15  | 3  | 40  | 60  | 22  | 14  | 4  |
| 56 | 59  | 21  | 14  | 6  | 55  | 59  | 20  | 14  | 6  | 60  | 59  | 21  | 15  | 5  |

Al, Zn < 1%

The measurements show that the electrolyte species silicon and sodium dominate the oxide coating composition. Only close to the substrate/layer interface there is more magnesium than silicon. Figure 10 shows a diffraction diagram of exemplary XRD measurements of untreated magnesium substrate and selected PEO coatings of the experimental plan. The diffractograms of the PEO layers show the same peaks like the untreated substrate and in addition a broad slightly peak between $2\Theta = 19–47^\circ$. This indicates that the peaks in the diffractograms originate from the substrate and the PEO layers by themselves have an amorphous structure. These observation are confirmed by experiments with the electrolytes of this experimental plan which were not part of the present study.

Figure 10. Diffractograms of the XRD analyses for the untreated AZ31 substrate and the PEO layers of experiment 6 (US=2, S=2, P=3, G=1) and 16 (US=2, S=3, P=3, G=2).

4. Summary and Conclusions

The influences of electrolyte composition and ultrasound pulses on the plasma electrolytic oxidation of the magnesium alloy AZ31 were investigated based on three-dimensional experimental plan according to the Box-Behnken design. Therefore, the electrical and optical process characteristics were observed and the PEO layer’s properties were examined. Within the investigated parameter range, the following findings were obtained.

The ultrasound pulses and phosphate concentration have no influence on the process voltage. The maximum anodic voltage was decreased by approximately 50 V by increasing the silicate concentration by 0.75 mol/l, which enables a lowering of the electrical energy consumption. Increased glycerol concentrations lead to a higher process voltage but also prevent the appearance of anodic voltage sags. The PEO coatings obtained in this study have an amorphous structure and consist of oxygen, silicon and magnesium mainly. Additionally, low amounts of sodium, zinc and aluminium are detectable. The atomic content of oxygen is around 60 % and does not vary in dependency of the
substrate distance. Close to the substrate ($d_{\text{substr.}} \leq 5 \, \mu\text{m}$) magnesium is in majority within the oxide structure with around 24% while in the outer parts of the coatings the magnesium silicon ratio is around 2:3. The high amount of electrolyte constitutes in the layer can be explained by the high concentrations of the used electrolytes. The coating method presented in this study is therefore a transitional form between a conversation and a deposition process. The modification of the electrolyte composition leads to different effects. Increased glycerol concentrations have a positive, statistically significant influence on all investigated layer characteristics. This could be explained by its process-stabilizing effect, which was suggested by the previously described influence on the course of the maximum anodic voltage. In general, glycerol can be identified as a beneficial electrolyte additive. The phosphate concentration has no statistically significant influence on the hardness, shows an increased layer thickness at level 2, and leads to a lowered corrosion current density at increased levels. Thus, phosphate is a statistically proven beneficial electrolyte additive for the plasma electrolytic oxidation of magnesium. However, the data obtained by this study give no clear explanation for its mode of action. There was no phosphorous detectable during the EDX analysis of the coating compositions. This suggests that the positive effect of increased phosphate concentrations can be justified by a modified layer morphology with a lower defect density. Increased silicate concentrations lead to thicker and harder coatings. This can be explained by the significant incorporation of silicon into the PEO layer. On average, the lowest and therefore best values for $i_0$ were obtained at the first and third silicate level. But two of the three experiments with the absolute lowest corrosion current density were obtained by experiments with a silicate concentration on level two, see table 3. This suggests that there are strong interactions between the electrolyte’s constitutes during the PEO-layer formation process. The assumption is supported by the resulting data of the optical process observation. The increase of the phosphate and glycerol concentrations lead to lower values for $i_0$ but has directly contradicting effects on the transition of discharge stage I into discharge stage II. Ultrasound has a strong influence on the duration of discharge stage I but no statistically significant effect on the layer thickness or corrosion current density. Its influence on the layer hardness is statistically significant but not in a technologically relevant magnitude.

A better understanding of the process parameter interactions could lead to an explanation of these partly contradictory observations. For a systematic statistical evaluation of such interactions the DOE used in this study is not appropriate. Therefore, selected areas of the parameter space should be investigated by full-factorial experimental plans. Additionally, the process monitoring used in this study could be improved: The measurements of the anodic and cathodic voltage maxima should be more high frequent. For a more effective evaluation of the data obtained by the optical process observation, automatized algorithms, which quantify discharge characteristics like duration, brightness and frequency are necessary.

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