THE INFLUENCE OF SOME REACTION CONDITIONS ON THE COURSE OF THE ELECTROLYTIC PREPARATION OF AMMONIUM PEROXODISULFATE

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As a continuation of the previous paper, the results of the examination on the influence of further operating parameters (anodic current density, anodic current concentration, molar ratio of the starting solutions, $P_0 = m^0_{H_2SO_4}/m^0_{(NH_4)_2SO_4}$, conversion degree, $\alpha = m_{(NH_4)_2S_2O_8}/m^0_{(NH_4)_2SO_4}$, composition and form of the anodic material, nature and concentration of anodic additives) on the anodic current efficiency χ and cell voltage of the laboratory flow-through integral electrolyzer model of the monopolar type were presented and discussed. The obtained results may be used for the optimization of the operating conditions of the industrial persulfate electrolyzer of the monopolar as well as bipolar type, too.

In our previous paper[1] the influence of reaction conditions on the course of electrolytic preparation of ammonium peroxodisulfate as the intermediate of the electrochemical production of hydrogen peroxide according to the Löwenstein's method was discussed. The results concerning the influence of the relative degree of saturation of starting solutions and the effect of temperature on the current efficiency χ and cell voltage of the laboratory flow-through integral monopolar electrolyzer were presented. The influence of other reaction parameters (anodic current density, anodic current concentration, molar ratio of the starting solution, $P_0 = m^0_{H_2SO_4}/m^0_{(NH_4)_2SO_4}$, conversion degree, $\alpha = m_{(NH_4)_2S_2O_8}/m^0_{(NH_4)_2SO_4}$, composition and form of the anode material and construction, nature and concentration of the anodic additives), originally planned for several following communications, were, however, not published due to the emigration of the main author of this research program. Although the industrial electrochemical production of hydrogen peroxide by the Löwenstein's process in the Czech Republic was replaced in the meantime by an autoxidation process, the electrosynthesis of ammonium peroxodisulfate did not loose its practical as well as theoretical importance as the initiator of various polymerization reactions and an oxidizing agent[2–4]. Therefore, the results of those investigations are presented now in a shortened form on the basis of the still available data. Their originality remained unaffected by other published papers in the related field[5–13].

EXPERIMENTAL

The laboratory model of the flow-through integral monopolar electrolyzer with one central anode compartment and two parallel cathode compartments separated by non-filtrating ceramic diaphragm plates as well as the measuring procedure and analytical methods were described in detail in the previous paper[1]. The composition of the starting solutions allowing to produce saturated ammonium peroxodisulfate in the anolyte at chosen operating conditions (P_0, α_{max} and anolyte temperature) were the same as in Table I of the previous paper[1]. The change in the anodic current concentration $j_{a,v}$ at the given anodic current density j_a was achieved by changing the current load I of the electrolyzer with simultaneous change of the total anode area and flow rate of the electrolyte, in order to keep the anodic current density and the desired conversion degree of the anolyte at the chosen value.

For the investigation of the influence of the composition and form of the anode material on the anodic current efficiency χ, the sheet strips or wires of pure platinum or platinum–iridium alloy with 5 wt.% Ir (Pt–Ir5) were used. The strips were 2 mm or 7 – 9 mm wide and 0.05 mm thick, the diameter of Pt wire was 0.3 mm, that of Pt–Ir5 wire was 0.2 mm.

RESULTS AND DISCUSSION

Influence of the Composition and Form of the Anodic Material on the Current Efficiency

The dependence of the anodic current efficiency χ on the anodic current density in the range of $j_a = 0.2 – 2.7$ A cm^{-2} for all used anodic materials, under the same reaction conditions, i.e. $P_0 = 1.5$, $\alpha = 0.8$, temperature of the anolyte 21.5 ± 0.5 °C, anodic current concentration $j_{a,v} = 27$ A dm^{-3} and without any anodic additives, showed distinct maxima at $j_a = 0.4 – 0.5$ A cm^{-2} for both sheet-strip materials, while for wire anodes of both compositions it was shifted to $j_a = 0.8$ A cm^{-2}. For platinum strips with different width varying from 2 to 9 mm no difference in the optimum current density was observed. These results have also shown that a little higher current yields may be obtained using wire anodes in comparison with sheet-strip anodes of pure platinum or Pt–Ir5 alloy. It followed further that about 3 – 5% higher current yields were achieved in the whole investigated range of current densities using anodes of pure platinum. In accordance with these results, all further investigations were done with anodes of pure platinum wires, irrespective to their lower corrosion and wear resistance in comparison with Pt–Ir5 alloy. Under such conditions it is possible to operate with a higher current load of the electrolyzer corresponding to a higher production capacity of the installed anode material in comparison with the sheet-strip anodes.

In order to explain the difference in the optimum current densities of the wire and sheet anodes of Pt and Pt–Ir5 the roughness factor of all investigated materials was determined by measuring the amount of adsorbed hydrogen. However, no distinct differences of this quantity were found ($f_r = 1.5 \pm 0.3$ for sheet as well as wire anodes). It was therefore assumed that the observed difference in catalytic properties is a result of the different technology of the sheet and wire fabrication, in accordance with the oxygen overvoltage on wire and sheet platinum anodes[14]. It is also possible that some influence on the shift of the maximum current efficiency to higher current densities for wire anodes may be caused by an enhanced heat exchange in comparison with the sheet-strip anodes, so that the true temperature at the surface of the wire anodes is lower than at the sheet-strip anodes. This temperature lowering is connected with the corresponding rise in the current efficiency[15–17].

Influence of Anodic Current Concentration on Current Yields

From the previous results[15–22] it is well known that the overall current efficiency of the electrolytic peroxodisulfates production is negatively influenced by preferential anodic discharge of monoperoxosulfate ions HSO_5^- formed in the anode compartment by spontaneous hydrolysis of $S_2O_8^-$ ions in acidic medium. In order to minimize the undesired amount of HSO_5^- ions in the anolyte it is necessary to keep the delay of the anolyte in the anode compartment as low as possible by operating at the highest anodic current concentration. However, the results of our measurements for $P_0 = 1.5$, $\alpha_{max} = 0.8$ or 0.4, $j_a = 0.6$ A cm^{-2} and cooling water temperature 20 °C, exhibited pronounced maxima of the current efficiency at $j_{a,v} = 27 - 35$ A dm^{-3} for both investigated values of the conversion degree. This theoretically unexpected finding of the maxima (a more distinct one for the higher conversion degree) can be explained in the following manner: the flow rate of cooling water was constant during all measurements, hence the increasing current load of the electrolyzer necessary for the increasing anodic current concentration leads to a temperature rise in the anodic compartment due to the enhanced Joule heat evolved there. The temperature increase causes the acceleration of the undesired hydrolysis of $S_2O_8^{2-}$ ions to HSO_5^- ions accompanied by the preferential anodic discharge of HSO_5^- to gaseous oxygen[19] decreasing in this way the overall anodic current efficiency of peroxosulfate ions. The rate of the anodic discharge of HSO_5^- ions is further enhanced by the increase in the transport rate of HSO_5^- ions to the anode surface due to the simultaneously increasing flow rate of the electrolyte through the anode compartment, which is necessary to held the conversion degree α at the desired constant level with increasing current load. At certain reaction conditions, however, the positive influence of the increasing anodic current concentration is just compensated by the negative influence of the increasing temperature in the anode compartment. With further increasing current load and flow rate of the electrolyte accompanied by the corresponding temperature increase in the anode compartment (e.g. at the highest current load the anolyte temperature is at 25 – 26 °C) their negative influence on the overall current efficiency becomes prevailing. With respect to the obtained results all further measurements were done at the constant anodic current concentration $j_{a,v} = 27$ A dm^{-3} corresponding to the optimum for the used electrolyzer.

Influence of the Anodic Current Density on the Current Efficiency without and with the Addition of Ammonium Rhodanide

Characteristic results for electrolysis without the addition of ammonium rhodanide are presented in Fig. 1. It shows the plot of the current efficiency χ vs anodic current density j_a for starting solutions with the maximum allowable concentration of educts with $P_0 = 1.5$ at 20 °C for the maximum attainable conversion degree $\alpha_{max} = 0.8$ and 0.4, respectively. The anodic current concentration was held constant at $j_{a,v} = 27$ A dm^{-3}. As may be seen, the course of this plot exhibits in both cases a distinct maximum at ca 0.75 – 0.8 A cm^{-2}, however, the current yields for the conversion degree $\alpha_{max} = 0.8$ (i.e. 80% of the original amount of $(NH_4)_2SO_4$ were converted to $(NH_4)_2S_2O_8$) are approximately 20% lower than those for the conversion degree $\alpha_{max} = 0.4$. Similar results were also obtained for starting solutions with other molar ratios P_0 and for other electrolysis temperatures. The maximum current efficiencies were shifted a little to the higher current densities with the increasing molar ratio P_0, thus $j_{a,opt} = 0.95 – 1.0$ A cm^{-2} for $P_0 = 2.5$ and $\alpha = 0.4 – 0.8$ at 10 – 30 °C.

The investigation of the influence of anodic additives on the optimalization of the electrolytic preparation of ammonium peroxodisulfate in the laboratory flow-through electrolyzer was only limited to ammonium rhodanide. According to our previous results[23–25] as well as results of other authors[3,5,7–9,11,12,15–17] it was repeatedly confirmed as the most suitable additive from other investigated substances of different types, with respect to its efficiency and price.

The results are shown in Figs 2 – 4. Figure 2 represents the plot χ vs feed rate of NH_4SCN, v, at three different current densities and all other reaction parameters held at

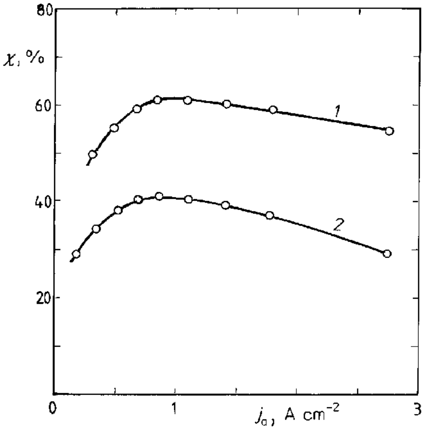

FIG. 1
Plot of χ vs j_a for starting solution with $P_0 = 1.5$, $\alpha_{max} = 0.4$ (*1*) and 0.8 (*2*), $j_{a,v} = 27$ A dm^{-3}, $t = 20$ °C, without anodic additives

the same level: $P_0 = 1.5$, $\alpha_{max} = 0.8$, $j_{a,v} = 27$ A dm^{-3} and cooling temperature 20 °C. All three curves exhibit distinct maxima of current yields which are shifted to greater feed rates with decreasing current density. This shifting of optimum feed rate is evidently caused by the longer delay of the anolyte in the anode compartment at lower current densities necessary to reach the same conversion degree α connected with the already discussed negative influence of the hydrolysis of peroxodisulfate ions to HSO_5^-.

As the change in the current density was accomplished by changing the number of anodic platinum wires, i.e. by changing the anodic surface at the constant current load, it seemed to be more suitable to correlate the influence of the feed rate of the additive against an uniform anodic surface area. The results from Fig. 2 recalculated in this manner are shown in Fig. 3. The maxima of the current yields for different current densities are achieved at about the same specific feed rate $\overline{v}$ of ammonium rhodanide corresponding to ca 1 mg (Ah)$^{-1}$ cm^{-2}. It means that the optimum specific feed rate of ammonium rhodanide is independent on the anodic current density.

Figure 4 represents the influence of the anodic current density on the current efficiency at three different feed rates of ammonium rhodanide for all other reaction parameters as given for Fig. 2. In contradiction to Fig. 1 for experiments without ammonium rhodanide addition, the maxima of current yields were obtained at ca j_a = 0.4 – 0.5 A cm^{-2} for all three feed rates. It means that the addition of NH_4SCN shifted the optimum anodic current density to lower values. About the same results were obtained also during measurements at other reaction parameters, P_0, α and t. Similar experiments have also shown that for our laboratory electrolyzer the optimum anodic

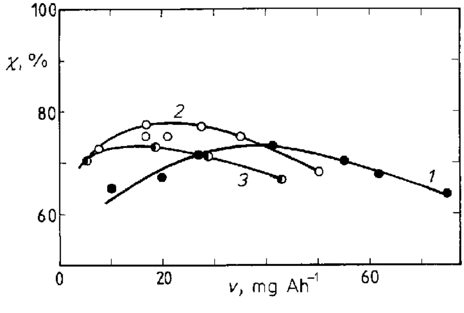

FIG. 2

Plot of χ vs feed rate of NH_4SCN, v [mg (Ah^{-1})] for j_a = 0.2 (*1*), 0.4 (*2*) and 0.8 (*3*) A cm^{-2} for starting solution with $P_0 = 1.5$, $\alpha_{max} = 0.8$, $j_{a,v}$ = 27 A dm^{-3}, t = 20 °C

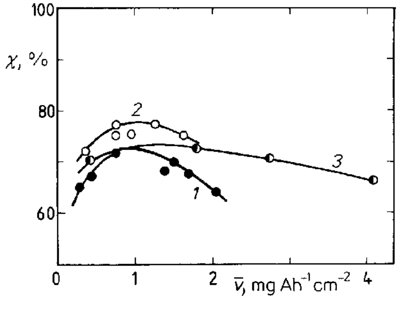

FIG. 3

Plot of χ vs specific feed rate of NH_4SCN, $\overline{v}$, [mg (Ah)$^{-1}$ cm^{-2}] (the same conditions as given for Fig. 2) current concentration during the experiments with ammonium rhodanide addition remained unchanged at $j_{a,v}$ = 27 – 30 A dm^{-3}.

Influence of the Conversion Degree α *and of the Molar Ratio* P_0 *of the Starting Solution on the Current Efficiency*

Some results about the influence of both these parameters were already shown in the previous paper[1] and also in Fig. 1. It can be concluded that the current efficiency decreases with increasing conversion degree α. Our results, however, were obtained using an integrally operating electrolyzer. In contradiction, the industrial electrolyzers representing an external or internal cascade of individual electrolytic cells, operate in a (quasi)differential regime. It is, however, well known that the current efficiency in an integral reactor is mostly lower in comparison to a cascade of (quasi)differentially operating cells for the same total conversion degree, especially for its high values. In order to obtain quantitative informations about these relations, the dependence of the current efficiency on the actual conversion degree α for starting solutions with maximum allowable conversion degrees α_{max} = 0.4, 0.6 and 0.8 and other reaction parameters P_0 = 1.0 – 2.5, t = 10 – 30 °C (Table I) for current densities j_a = 0.4 – 2.7 A cm^{-2} without and with addition of ammonium rhodanide was investigated. It is not possible, however, to present here all results of these measurements. Therefore, only two characteristic examples are shown in Fig. 5 and 6. Figure 5 depicts the plot χ vs α for electrolysis of the starting solutions with P_0 = 1.0 – 2.5, α_{max} = 0.8, cooling temperature t = 20 °C, anodic current density j_a = 2.7 A cm^{-2}, anodic current concentration $j_{a,v}$ = 27 A dm^{-3}, without ammonium rhodanide addition. As expected, a monotonous decrease of the

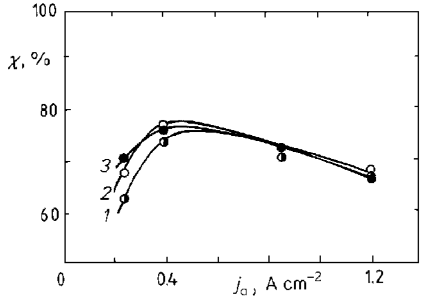

FIG. 4

Plot of χ vs j_a for different feed rates of NH_4SCN: v = 10 (*1*), 20 (*2*) and 30 (*3*) mg $(Ah)^{-1}$. Other reaction conditions are the same as given for Fig. 2

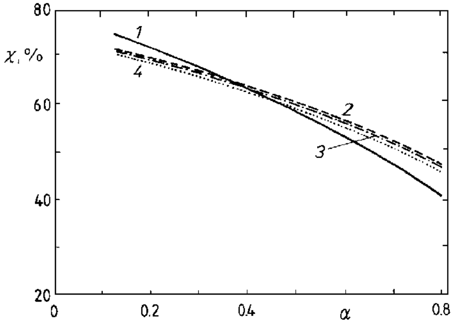

FIG. 5

Plot of χ vs actual conversion degree α for starting solutions with various molar ratio P_0 = 1.0 (*1*), 1.5 (*2*), 2.0 (*3*) and 2.5 (*4*), α_{max} = 0.8, t = 20 °C, j_a = 2.7 A cm^{-2}, $j_{a,v}$ = 27 A dm^{-3}, without anodic additives

TABLE I
Composition of the applied starting solutions of maximum allowable concentration[32]

P_0	Starting component wt.%	10 °C			20 °C			30 °C		
		α_{max}			α_{max}			α_{max}		
		0.4	0.6	0.8	0.4	0.6	0.8	0.4	0.6	0.8
1.0	H_2SO_4	17.99	17.41	17.27	19.73	19.29	19.05	21.41	20.76	20.58
	$(NH_4)_2SO_4$	24.55	23.47	23.28	26.86	25.99	25.68	28.86	27.97	27.71
	m_T[a]	6.466	6.008	5.926	7.612	7.188	7.032	8.784	8.258	8.110
1.5	H_2SO_4	21.88	21.08	20.63	24.22	23.18	22.65	26.30	25.07	24.56
	$(NH_4)_2SO_4$	19.65	18.94	18.53	21.77	20.82	20.34	23.62	22.52	22.07
	m_T[a]	6.358	5.974	5.762	7.626	7.034	6.750	8.923	8.129	7.824
2.0	H_2SO_4	24.90	23.87	23.18	27.46	26.27	25.54	30.10	28.44	27.60
	$(NH_4)_2SO_4$	16.78	16.08	15.62	18.50	17.70	17.21	20.29	19.17	18.59
	m_T[a]	6.532	6.079	5.795	7.772	7.172	6.825	9.285	8.307	7.843
2.5	H_2SO_4	27.57	26.11	25.55	30.37	28.61	28.08	33.25	31.12	30.15
	$(NH_4)_2SO_4$	14.90	14.07	13.75	16.36	15.42	15.15	17.95	16.77	16.25
	m_T[a]	6.860	6.230	6.000	8.135	7.297	7.069	9.743	8.524	8.030

[a] Total solution molality $m_T = (P_0 + 1)\, m_{(NH_4)_2SO_4}$ (in mol $kg^{-1}_{H_2O}$).

current efficiency with increasing actual conversion degree was observed for all starting solutions. The current yield values were decreasing with growing molar ratio P_0 of the starting solution. Similar results were obtained for most measurements with varying reaction parameters. Some exception could be observed during the measurements with the starting solutions for the electrolysis at 10 °C, where the decrease of the plot χ vs α for $P_0 = 1.0$ was steeper than for higher values of this parameter (see Fig. 6). This irregularity could not be explained satisfactorily.

The decrease of the current efficiency with rising actual conversion degree is caused first of all by the decreasing concentration of SO_4^{2-} ions as the main discharging particles for the electrosynthesis of peroxodisulfate ions[26] so that the partial current density of their discharge diminishes and, consequently, the partial current density of the second main anodic process, oxygen evolution, increases at the given total anodic current density in the galvanostatic regime. Some fraction of the current efficiency decrease is caused by the longer delay of the anolyte in the anode compartment leading to a correspondingly larger amount of the hydrolytically formed HSO_5^- ions[21,22] and their preferential discharge to gaseous oxygen[15–17,19,20].

The usually observed decrease of the current efficiency with increasing molar ratio P_0 of the starting solution at α unchanged is caused mainly by the decreasing amount of SO_4^{2-} ions as the main discharging particles in the formation of $S_2O_8^{2-}$ (refs[26,27]). Secondly, the concentration of H^+ ions in the anolyte increases with the increasing value of P_0. It catalyses the undesired hydrolytic decomposition of $S_2O_8^{2-}$ to HSO_5^- in the anode compartment, leading to the enhanced anodic evolution of gaseous oxygen.

The previous conclusion about a much greater discharge rate of SO_4^{2-} ions in the $S_2O_8^{2-}$ formation in comparison with the discharge rate of HSO_4^- ions[26] confirmed by the present results may further be supported by Fig. 7 in which the results from Fig. 5 were transformed into the plot χ vs α' where $\alpha' = 2\alpha/(P_0 + 1)$ denotes the actual conversion degree related to the sum of both dischargeable anions, SO_4^{2-} and HSO_4^- . If there was

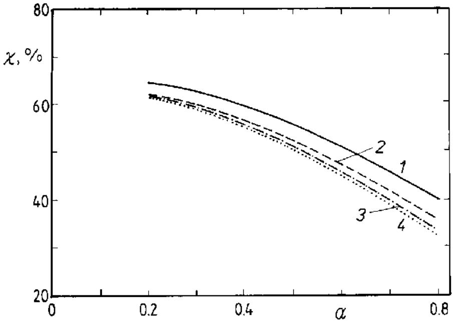

FIG. 6

Plot of χ vs actual conversion degree α for starting solutions with various molar ratio P_0 : 1.0 (*1*), 1.5 (*2*), 2.0 (*3*) and 2.5 (*4*), $\alpha_{max} = 0.8$, $t = 10$ °C, $j_a = 2.7$ A cm^{-2}, $j_{a,v} = 27$ A dm^{-3}, without anodic additives no substantial difference in the specific discharge rate of SO_4^{2-} and HSO_4^- anions leading to $S_2O_8^{2-}$ then, due to the fact that the summary content of the S-containing anions in the starting solutions is almost the same in the whole range of P_0 = 1.0 – 2.5 (Σm = 6.75 – 7.07 mol $kg_{H_2O}^{-1}$ for α_{max} = 0.8 and t = 20 °C, see Table I), no substantial difference in the current yields could be expected for the same value of the actual conversion degree α' for the starting solutions with the different value of P_0. In reality, however, a very distinct difference was obtained for the current yields at the same actual conversion degree α' for different values of P_0 (see Fig. 7). A clear decrease of the current yields with the increasing molar ratio P_0 and unchanged value of α' stays in a full accordance with the decreasing concentration of SO_4^{2-} ions in anolyte.

Figure 8 shows the dependence of the cell voltage on the molar ratio of the starting solutions with α_{max} = 0.8 for electrolysis at 10 °C and for various values of their actual conversion degree α = 0.2 – 0.7, too. The greatest difference in the cell voltage of the laboratory monopolar electrolyzer exists for the molar ratio P_0 = 1.0 with increasing actual conversion degree α. The voltage difference decreases with increasing P_0 of the starting solution. As expected, the lowest cell voltage values were in all cases attained for electrolysis of the starting solution with the highest value of P_0, i.e. with the highest concentration of H^+ ions in the electrolyte. It results in the highest conductivity and correspondingly lowest ohmic voltage drop between both electrodes[28]. Similar results were obtained for starting solutions with other values of the reaction parameters (α_{max}, t, j_a, anodic additives etc.).

Finally, it can be concluded that the measurement results of this communication, representing only a characteristic selection of all similar experimental data for all

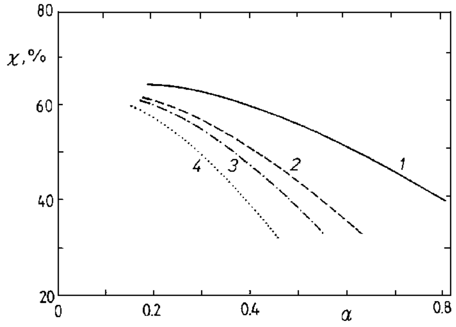

FIG. 7

Plot of χ vs actual conversion degree α' for starting solutions with various molar ratio P_0 = 1.0 (*1*), 1.5 (*2*), 2.0 (*3*) and 2.5 (*4*), α_{max} = 0.8, t = 20 °C, j_a = 2.7 A cm^{-2}, $j_{a,v}$ = 27 A dm^{-3}, without anodic additives

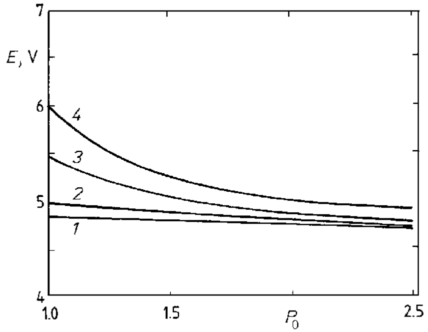

FIG. 8

Plot of the total cell voltage E vs molar ratio P_0 of starting solutions with α_{max} = 0.8, at different actual conversion degrees α = 0.2 (*1*), 0.4 (*2*), 0.6 (*3*) and 0.7 (*4*), j_a = 2.7 A cm^{-2}, $j_{a,v}$ = 27 A dm^{-3}, t = 10 °C, without anodic additives possible values of individual reaction parameters, can be used simultaneously with the results of the previous paper[1] for the optimization of the process under study realized in the monopolar as well as in the bipolar electrolyzer[2–4,15-17,29–31].

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