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Statistical analysis on the variables affecting itaconic acid separation by bipolar membrane electrodialysis

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ABSTRACT

Itaconic acid (IA) is a well-known, unsaturated, dicarboxylic acid. It is widely used in plastic industry as a co-polymer. Nowadays the itaconic acid is mainly produced by fermentation. Due to the product inhibition effect during the batch operation, the continuous/semi-continuous separation of itaconic acid from the fermentation broth is highly desired. The electrodialysis seems to be a promising alternative. The effect of three parameters, namely initial itaconic acid concentration, pH, and applied potential on the efficacy of the electrodialysis were studied. More than 90% of current efficiency could be achieved by changing of the main factors mentioned above. From the results it is clear, that electrodialysis with bipolar membrane is an adequate method for the separation of itaconic acid.

Keywords: Electrodialysis; Itaconic acid; Current efficiency; Bipolar membrane; Integrated bioprocess

1. Introduction

Itaconic acid was discovered by Baup in 1837 [1]. The biochemistry of the production of itaconic acid is rather like the evolves of citric acid. Itaconic acid (2-methylene,1,4-butanedioic acid) is an unsaturated dicarboxylic acid, which has two carboxylic group and a conjugated double bond. In fact, it can be utilized as a chemical building block because of its beneficial chemical properties (Table 1). It can be applied as raw material in plastic industry [2]. Thanks to the increasing importance of sustainable components, IA has gained more and more interest.

Itaconic acid can be produced by fermentation with several strains such as *Aspergillus itaconicus* [4], *Ustilago maydis* Steiger [5], *Pseudozyma antarctica* [6], *Yarrowia lipolytica* [2], *Aspergillus terreus* [7], *Synechocystis cyanobacteria* [8,9]. The industrial production of IA occurs entirely by submerged fermentation with the fungus *A. terreus* [10] and is one of the major examples of organic acid production by *Aspergillus* spp. thanks to metabolic overflow of primary metabolism. Nemestóthy et al. [11] investigated the IA fermentation by *A. terreus* and followed the fermentation process by CO_2 measurements.

To increase the production of itaconic acid, there are two main directions: increase the efficiency of fermentation and product recovery. During the fermentation, it seems to be key parameters are temperature, pH [12], aeration [13]. Karaffa et al. [10] found, that effect of manganese ion had main role in achieving high yield of itaconic acid. In industrial scale the product recovery takes place by adsorption on activated carbon and repeated crystallization. The steps of the classical (sequential) recovery of itaconic acid and

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Table 1
Physical and chemical properties of itaconic acid [3]

Description	White, crystalline, monounsaturated organic diacid
Formula	$C_5H_6O_4$
Molar mass	130.1 g/mol
Solubility in water (20°C)	83.1 g/L
Melting point	167°C–168°C
Boiling point	268°C
Density (25°C)	1.573 g/cm^3
Protonation states	H_2Ita, HIta$^-$, and Ita^{2-}
Dissociation constants in aqueous solutions (at 15°C–45°C)	3.83–3.89 (pK_{a1})
Dissociation constants in aqueous solutions (at 15°C–45°C)	5.41–5.46 (pK_{a2})

related cleaning processes could be reduced and simplified by the system integrating fermentation and electrodialysis with bipolar membranes [14]. The application of electrodialysis can reduce the product inhibition effect during the fermentation process, too [15]. Stodollick et al. [16] gave a mathematical model for electrodialysis with bipolar membrane. Varga et al. [14] tried to find an answer, whether electrodialysis can be a suitable method for the recovery of itaconic acid. There are limited publications and information in the field of itaconic acid separation by EDBM so far.

The novelty of this study is the use of statistical method to determine key-factors having with significant effect on itaconic acid separation by a commercialized electrodialysis unit. The results could bring closer the possibility of integrated system design/operation connecting the fermentation and the electro-membrane technique. In this study, the impact of three main parameters (initial itaconic acid concentration, pH, and applied potential of electrodialysis) was investigated employing model solutions. During the experiments, electric conductance, pH, and current were registered and were looked for the correlation between the main parameters and current efficiency. Itaconic acid concentration was measured in the segments of electrodialysis unit to follow the separation and migration of the components. In a previous study, Varga et al. [14] applied monopolar membranes. Bipolar membranes were tested to increase the capacity of the unit. With the application of bipolar membranes, cation- and anion-exchange membranes in three different segments were applied (shown in Fig. 1), one for the collection of initial model solution (C2) and one for the separated sodium (C1) and one for the itaconate ions separated (D2). The separated sodium hydroxide can be utilized to control the pH during the itaconic acid fermentation process.

2. Materials and methods

2.1. Electrodialysis with bipolar membrane

The components of itaconic acid fermentation broth are mainly glucose, maltose, itaconic acid, malic acid, alpha-ketoglutaric acid, fumaric acid, and some metal containing molecules in much lower concentration [17]. As glucose and maltose are neutral molecules in the fermentation broth and IA in dissociated form (mono-, or di-itaconate, depending on pH), the application of electrodialysis unit seems to reasonable to remove of IA from the fermentation liquor [14,16].

In this study the P EDR-Z/4x unit (made by MemBrain®) functioned as EDBM-Z/4x10-0.8 Type CAB method which contained the series of anion exchange-, cation exchange-membranes as a classical electrodialyser and bipolar membranes, too. At the ends of the ED unit there were platinum coated electrodes, which connected with positive and negative pole of the power supply. The electrodes were surrounded by solution of sodium sulphate as electrolyte. Among the electrodes 10 segments of cation- and anion-exchange and bipolar membranes were built in to increase the capacity of the separation. The schematic draw of the equipment can be seen in Fig. 1.

The model solution was fed (1 L) into the C2 segment and due to the electric force, sodium ions, and itaconate ions moved to the direction of negative and positive pole, respectively. The glucose as neutral molecules remained in the C2 segment. The sodium ions could migrate through the cation-exchange membrane and the itaconate ions through the anion-exchange membrane. The bipolar membrane functioned as a two-sided electrode to split the water molecules (OH^-, H^+). At the beginning C2 tank contained itaconic acid solution, C1 and D2 tank was filled with 1–1 L of deionized water. Finally, the itaconate ions were collected in D2 segment, the sodium ions in C1 segment and the glucose in C2. The solution of the segments was collected in the C1, C2, D2 buffer tanks, respectively. Between the segment and buffer tank, each solution was circulated separately by pumps. The electrolyte was collected in tank E (D1 segment was not used in this study). The experimental electrodialysis equipment can be seen in Fig. 2.

2.2. Materials and test conditions

The model solution was itaconic acid (CAS No.: 97-65-4, Acros Organics™, New Jersey, USA: 1-800-ACROS-01, purity: >99%) solution. The pH was adjusted by sodium hydroxide. The electrolyte was 0.5 mol/dm^3 sodium sulphate solution. Sodium hydroxide, sodium sulfate, sulfuric acid, and all other chemicals were analytical grade and purchased from Sigma-Aldrich, (3050 Spruce Street, Saint Louis, MO 63103, USA).

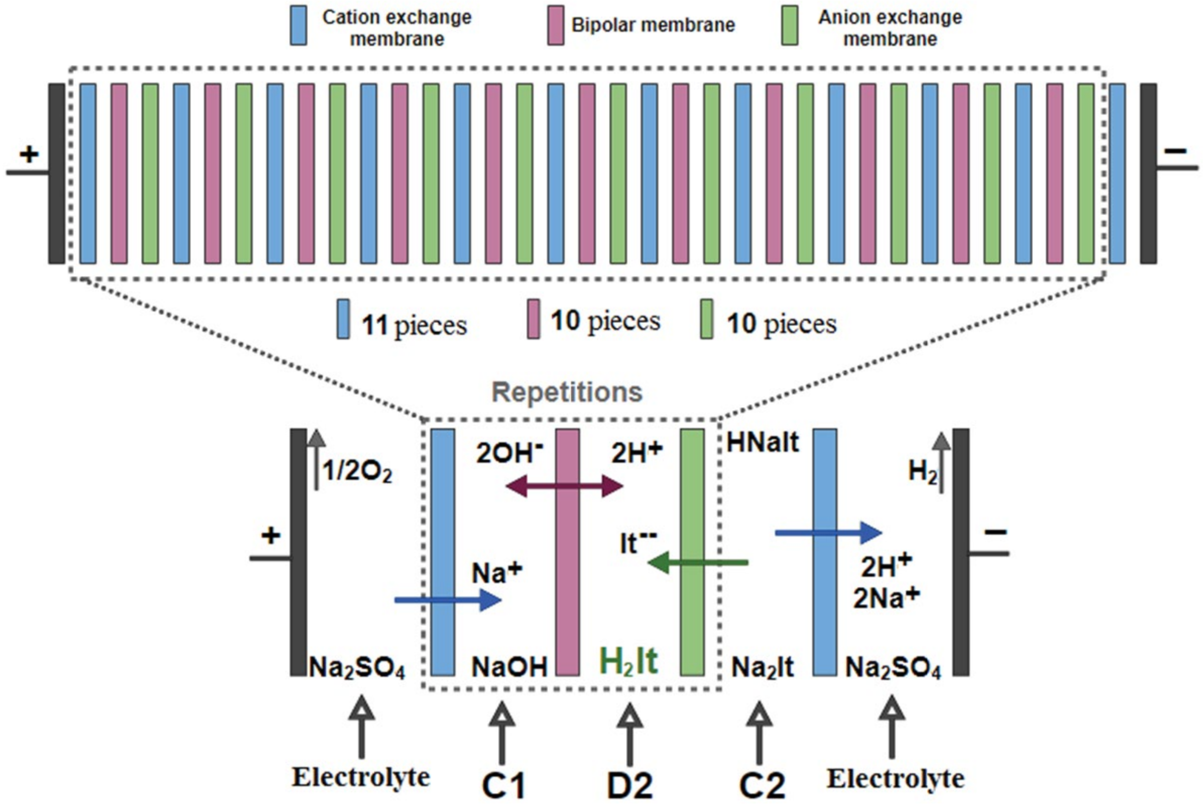

Fig. 1. Scheme of the membrane module in the electrodialysis equipment and the predominant transport mechanisms during the separation.

Fig. 2. Image of the multifunctional P EDR-Z/4x electrodialysis equipment.

The dissociation of itaconic acid depends on the pH, as illustrated in Fig. 3.

The P EDR-Z/4x electrodialyser equipment contained RALEX EDR-Z type membranes. The main parameters of the equipment are summarized in Table 2. The operating flow rates were set in all cases as follows: C1: 50 L/h, D2: 50 L/h, C2: 50 L/h, and E: 60 L/h.

For analytical purposes a Young Lin Instrument Co., Ltd., (899-6 Hogye-dong, Anyang, 431-836, Korea) (YL9100-type) high-performance liquid chromatography (HPLC) system (including a YL9109 vacuum degasser, YL9110 quaternary pump and YL9150 automatic sample dispenser) was used to determine the concentration of itaconic acid with a Hamilton PRP-X300 HPLC column (25 cm in length, 4.1 mm inner diameter, 7 µm particle size) and a YL9120 UV/Vis detector.

The electric conductance of the solution of the segments was measured by a Radelkis OK-102/1 conductivity meter equipped with a Radelkis OK-9023 bell electrode using a cell constant of 0.7 cm^{-1}. The data were collected online using LabVIEW software. The pH of each segment was measured by a Radelkis OP-205/1 pH meter equipped with WTW SenTix 60 type electrode. The current was measured online by Metrix MTX 3281 multimeter and the data were evaluated by SX-DMM v 2.3 software.

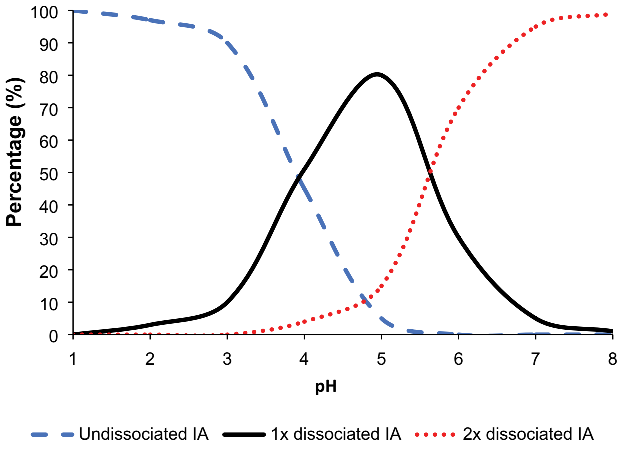

Fig. 3. Dissociation of itaconic acid depending on pH [11].

2.3. Experimental plan

The effects of three parameters on the current efficiency of the electrodialysis were investigated. These were: initial itaconic acid concentration of C2 solution (1 L), initial pH of C2 solution and applied voltage of electrodialysis. The studied range was determined based on fermentation data/feedback, for example, in terms of IA concentration and pH as well as the features of the electrodialysis equipment, for example, in terms of adjustable potential. Accordingly, the initial IA concentration of C2 solution was ranged between 5 and 60 g/L, the initial pH of C2 was varied between 2 and 8, while the applied voltage was changed between 10 and 30 V. The 5–60 g IA/L is a realistic concentration range in the fermentation broth to be reached relative easily in a typical batch [18]. The general pH of the fermentation is between 2 and 3 [17,18]. In this work, the pH range was expanded from 2 to 8. Thanks to the wide pH range, the various dissociated forms of itaconic acid could be investigated in the course of the separation (seen in Fig. 3). The impacts of these variables were analyzed by experimental design, which is an approach that can provide reliable information about the significant factors (affecting the ED current efficiency as output/response parameter) and support therefore the validity of conclusions thanks to its mathematical statistical background referred as ANOVA (analysis of variance) [19]. In this work, a 2-level, 3 factorial experimental design was chosen with repetitions in the centre point (C) and curvature check (Table 3).

The actual current data through the electrodialysis unit was collected as a function of time. The current efficiency ($\eta_{current}$) was calculated as follows:

Table 2
EDBM-Z/3x10-0.8 Type CAB module specification and operating limits

Parameter	Value
Effective surface of the membrane module	1,984 cm^2
Effective surface of one membrane in the module	64 cm^2
General size of membranes	56 mm × 206 mm
Effective size of membranes	40 mm × 160 mm
Number of the membrane cells	10 pieces
RALEX anion exchange membrane AM(H)-PP	10 pieces
RALEX cation exchange membrane CM(H)-PP	11 pieces
RALEX bipolar membrane (PP)	10 pieces
Thickness of the spacer	0.8 mm
Electrode (anode, cathode), Ti/Pt	2 pieces
Size of ED module	135 mm × 90 mm × 250 mm
Weight of module with preservation solution	1.9 kg
Operating flow-rates	25–60 L/h
Minimum and Maximum temperature	10°C–35°C
Operating voltage	3 V/triplet
Maximum voltage/current	35 V/3 A

Table 3
Summary of the parameters of the experiments and the resulted current efficiency

Initial IA concentration (g/l)	pH	Applied voltage (V)	Current efficiency (%)
5	2	30	69.0
5	8	10	80.4
33	5	20	96.6
33	5	20	99.2
33	5	20	91.9
60	2	10	88.4
60	8	30	66.1

$$\eta_{\text{current}}(\%) = 100 \times \frac{Q_{\text{theoretical}}}{Q_{\text{eff}}} \tag{1}$$

$$Q_{\text{theoretical}} = E_m \times b \times F \tag{2}$$

$$Q_{\text{eff}} = \int I \times dt \times N \tag{3}$$

where in the case of $Q_{\text{theoretical}}$ (2) the E_m is the total mass equivalent of itaconic acid which theoretically transported (mol), b is 2 for fully dissociated itaconic acid, F is the Faraday constant (96,500 As/mol). In the case of Q_{eff} (3) N is the number of cell pairs (in this study it is 10), I is the recorded current intensity (A), and t is the duration of the itaconic acid separation (s) [20].

3. Results and discussion

3.1. Evaluation of the ED process

During electrodialysis experiments, three parameters were recorded continuously and analyzed afterwards. These data were as follows: pH and conductance of each segment (C2, C1, D2) and the current through *t* module. The electric potential ensured by the DC supply was fixed during a given experiment.

The first step in this study was the control of efficiency of the electrodialysis unit. The series of experiments were run 33 g/L of initial itaconic acid in C2 (shown in Figs. 1 and 2) and IA concentration in D2 was recorded in time. The IA concentration was determined by HPLC. The change of the IA concentration is presented in Fig. 4. At the end of the electrodialysis, the itaconic acid concentration was measured in each segment. It was found, that more than 90% of the initial itaconic acid was in D2 container. This result shows relatively high selectivity of the system.

The main results, namely the time dependent current (*A*), the pH (*B*), and the conductance of segments (*C*) are presented in Fig. 5. These types of graphs were made in each experiment and were considered to draw conclusions.

In Fig. 5a, a typical current curve is shown. The overall current flowed through the module. The current was registered by the same equipment with different conditions. The initial condition of tests is summarized in Table 3. In each cases the overall current has a maximum. This is due to the alteration of the resultant resistance of the segments in time. The three segments, electrically, can be regarded as three resistors connected in series. At the beginning, D2 and C1 segment were filled with 1–1 L of deionized water, which had high resistance (resulted in low current). During the separation, each segment contained ions thanks to ion migration, which resulted in lower resultant resistance (higher current). The segment of C2 became steadily free of mobile ions, therefore at the end of separation, the resultant resistance increased again (lower current).

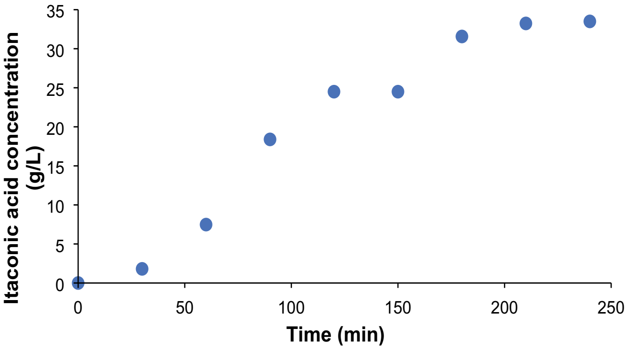

Fig. 4. Time profile of itaconic acid enrichment in D2. Experimental conditions as given in the third row of Table 3.

In Fig. 5b can be seen a typical pH curve during the electrodialysis. As it was expected, the pH increased in C1. The sodium ions migrated in this direction. In D2 the pH decreased considerably, due to acidic character of migrating itaconic acid. At the end of the experiment, in C2 the pH was near to neutral, because sodium and itaconate ions moved to C1 and D2, respectively. In Fig. 5c, a typical electric conductance curve can be seen. The conductance was registered by the same equipment with different conditions. The initial conditions are summarized in Table 3. In the case of C2 segment, the conductance decreased due to the migration of itaconate and consequently lower concentration of ions. The conductance in the D2 and C1 segments had strong correlation with pH and current curves. The appearance of H^+ and OH^- led to higher conductance. Each experiment was characterized based on the current efficiency. The current efficiency was calculated using Eqs. (1)–(3). The value of Q_{eff} was determined from the integral of the current vs. time curve (area under the curve). The time demand of the experiment was longer if the initial pH was low (low initial Na^+ content) and/or if the applied electric potential was low. In all cases, the conductance of C2 declined to the level deionized water. The final conductance of D2 and C1 depended on the initial concentration of itaconic acid and sodium ion in C2 solution. These ions migrated from C2 to D2 and C1, respectively. The current efficiency was determined from the integral of the current vs. time curve (area under the curve).

From the results (summarized in Table 3.), it can be seen that, from the initial itaconic acid concentration point of view, it was beneficial to apply 33 g IA/L to improve the current efficiency of electrodialysis. For the determination of optimum, further experiments are needed.

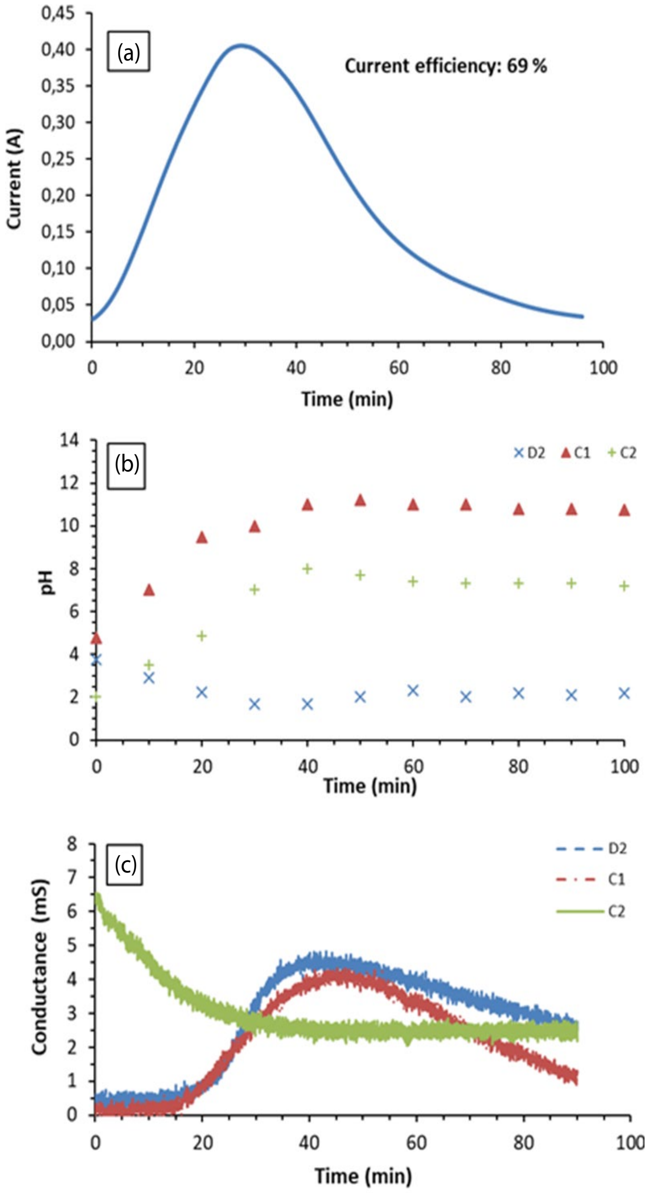

Fig. 5. Example for the change of (a) current, (b) pH, and (c) solution conductance during electrodialysis. Experimental conditions as given in the first row of Table 3.

On the one hand, it is clear that the electrodialysis could be driven more rapidly by applying higher electric potential and/or pH (higher electric conductance of the solution). On the other hand, however, the higher potential can lead to a critical current that can decrease the current efficiency of the itaconic acid separation. In previous work Varga et al. [14] achieved promising results with at low IA content (3 g/L) model solution by a conventional ED system. Tongwen and Weihua [21] obtained >70% current efficiency. They observed that the actual current efficiency was dependent on time. Moresi et al. [22,23] achieved average 61% and later, in 2010, more than 96% current efficiency, which is a quite similar value to those reported in this work. Prochaska and Woźniak-Budych [24] separated fumaric acid by bipolar membrane electrodialysis.

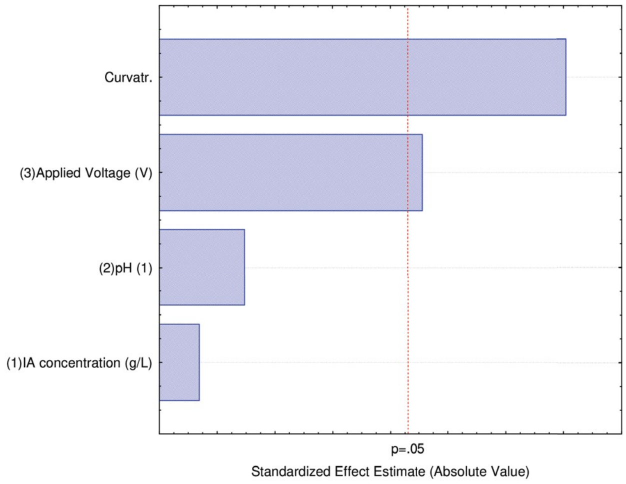

Fig. 6. Pareto-diagram demonstrating the results of ANOVA.

Fumaric acid (FA) has similar properties than itaconic acid. In their work under different current densities (90, 120, 150 A/m^2) and 50%–90%, 20%–85% current efficiency, product recovery were achieved, respectively with relatively low FA concentration (1.45–2.90 g/L). Lameloise and Lewandowski [25] recovered L-malic acid (also similar to IA) from a beverage industry waste water by two-compartment bipolar electrodialysis. Acid recovery was in the range of 93%–97% while CE was between 87% and 97%. Otherwise malic acid as one of the main by-products is generated during the IA fermentation [17].

3.2. Results of experimental design and ANOVA

The values of current efficiency obtained under various ED operating settings can be observed in Table 3.

The results of ANOVA conducted on the data of Table 3 are illustrated in the Pareto-chart (Fig. 6), where it is to observe that among the input variables, only electric potential applied was found to have a statistically relevant effect ($p < 0.05$).

For instance, the 3-D response surface considering the applied voltage and initial IA concentration is displayed in Fig. 7. It can be drawn, in agreement with the Pareto-chart (Fig. 6), that IA concentration had negligible influence, while under lower applied voltages, the current efficiency of the ED process could be enhanced. This conclusion can be explained by electrochemical background of electrodialysis (e.g., different migration speed of the ions).

Furthermore, it is indicated by the $p < 0.05$ value of curvature check in ANOVA that the effect of applied voltage needs further elaboration since a linear model may not be sufficient to describe its relationship with ED current efficiency. This should be studied in a future research in order to find the optimal parameter settings of the separation process.

4. Conclusion

In this study we investigated the effect of initial itaconic acid concentration, pH and the voltage on the efficiency of electrodialysis. It is obvious, that this technique is available

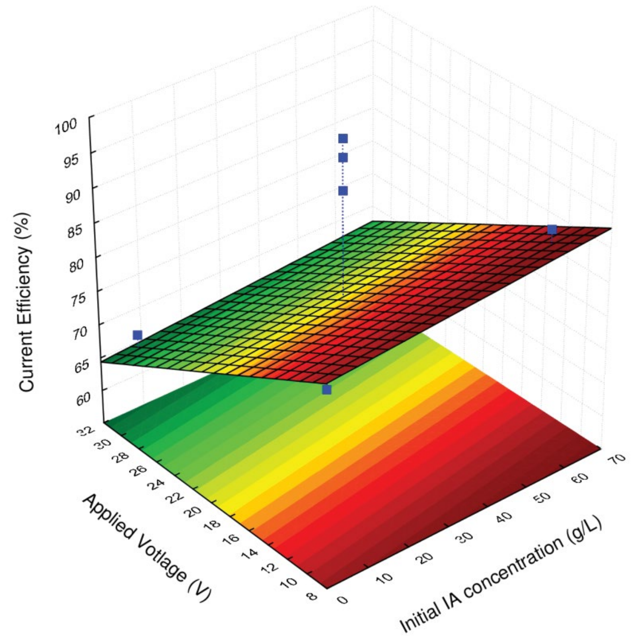

Fig. 7. Dependency of current efficiency on applied voltage and initial IA concentration.

for the separation of itaconic acid. With the changing of three parameters the process can be optimize and we can achieve high current efficiency (99.2%). Based on the results we can conclude, that electrodialysis is able to separate itaconic acid from model solutions. The next step is to analyse more complex mixture especially the real fermentation broth to find out how can we selectively separate itaconic acid. If it is successful, we will be able to design an integrated process where the product formation and recovery can be performed simultaneously.

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