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Recovery of alpha-ketoglutaric acid from multi-component model solutions: impact of initial composition of diluate solution on the efficiency of the EDBM process

Mateusz Szczygiełda, Krystyna Prochaska*

Institute of Chemical Technology and Engineering, Poznan University of Technology, Berdychowo Street 4, 60-965 Poznań, Poland, Tel. +48 61665 3601; Fax: +48 61 665 3649; email: krystyna.prochaska@put.poznan.pl (K. Prochaska)

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ABSTRACT

In this work, the impact of an initial ratio of concentrations of alpha-ketoglutaric acid (AKG) in relation to lactic, acetic, and propionic acids present in a multi-component simulated solution, on the efficiency of the bipolar membrane electrodialysis (EDBM) process as well as on the purity of main product was investigated. The use of 10 cell pairs EDBM membrane stack with a membrane configuration of anion-exchange membrane–bipolar membrane allowed for the total removal of AKG from diluate solution at a constant current density (90 A/m^2) in less than 90 min. Moreover, an increase in the initial concentration of AKG in diluate solution from 15 to 25 g/L was found to lead to a significant improvement of the purity of AKG obtained, from 58.2% to 80.6% at the end of the optimal duration of EDBM process, at the same specific energy consumption (1.8 kWh/kg).

Keywords: Alpha-ketoglutaric acid; Acid recovery; Electrodialysis; Bipolar membrane electrodialysis

1. Introduction

The bipolar membrane electrodialysis (EDBM) is one of the electromembrane techniques, which can be effectively used in the processes of brine desalination, waste treatment, whey electroacidification as well as organic acids recovery [1–4]. The presence of bipolar membrane (BP) in the membrane stack, the main role of which is water splitting (H^+ and OH^- ions production) at an applied electric field, allows for environment-friendly conversion of salts to the corresponding bases and acids [5]. It is especially important in the process of recovery of organic acids from the actual post-fermentation broth due to the possibility of elimination of acidification step and pollution reduction [6,7]. In our study, alpha-ketoglutaric acid (AKG) was selected as the main product of fermentation of carbon sources due to the wide range of its applications in medicine and pharmacy such as: dietary supplement, important source of amino acids for collagen synthesis, and as a component for wound healing [8–10]. It is worth to mention that in the processes of recovery of organic acids from the actual post-fermentation broth, the use of proper membrane stack configuration enables partial separation of ionic/dissociated forms from nonionic/nondissociated compounds [11]. However, the selective separation of a proper ionic component from other carboxylic acids included in the multi-component mixture is difficult to achieve. Consequently, the transport of by-products leads to the increase in the cost as well as to the decrease in the process efficiency and purity of the product obtained. For example in 2012, Lameloise et al. [12] successfully used the EDBM process as one of the step of l-malic acid recovery from other by-products of the alcoholic fermentation process. However, they indicated that the presence of organic by-products (acetate, lactate, and citrate) in real broth led to a significant increase in a specific energy consumption (50% higher), when compared

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* Corresponding author.

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with a one-component simulated solution. One of the main factors that may significantly affect the efficiency and selectivity of the process is the initial ratio of concentrations of the main product to by-products present in the diluate solution. A potential strategy is to control the concentration of compounds obtained in the fermentation process as well as in the pre-treatment before EDBM process. Hence, in order to optimize the EDBM process before the recovery of AKG from the actual post-fermentation broth, the studies with the use of simulated solutions were performed. The impact of the initial ratio of concentrations of AKG to lactic (LA), acetic (AA), and propionic (PA) acids present in the simulated solutions, on the efficiency of the EDBM process as well as on the purity of the main product was investigated.

2. Material and methods

2.1. Materials

In this study, two multi-component simulated solutions (further noted as simulated broths) with different initial concentrations of AKG (*Alchem,* Poland) equal to 15 and 25 g/L were used as the diluate solutions in EDBM process. In addition, both simulated broths contained such components as: LA, 10 g/L; AA, 5 g/L; PA, 2.1 g/L; glucose (Gluc), 2.1 g/L; and ethanol (EtOH), 0.7 g/L (*Sigma-Aldrich*, Poland) and were prepared using distilled water of conductivity not-exceeding 3 μS/cm. The composition of simulated solutions was determined on the basis of the composition of the actual post-fermentation broth delivered by the Poznan University of Life Sciences (Poland). Sodium alpha-ketoglutarate solution, which was fed to concentrate compartment, was prepared by mixing the aqueous solution of AKG with NaOH (*POCH,* Poland). Moreover, sulfuric acid (96%) (*Sigma-Aldrich*, Poland) was used to prepare electrolyte solutions as well as eluents for the high-performance liquid chromatography (HPLC) analysis.

2.2. Membranes

Commercially available anion-exchange membranes (AMs) (*PC 200D*), a cation-exchange membrane (CM) (*PC-SK*), and BPs (*PC bip*) by *PCCell GmbH* (*Germany*) were used in this study. The main properties of applied ion-exchange membranes are presented in Table 1.

2.3. EDBM experiments

In our study, EDBM batch system equipped with AM–BP stack configuration was selected in order to simultaneously remove AKG anions from multi-component simulated solutions and to convert them to the acidic form. 10 AMs and 10 BPs (which were separated by spacers of 0.5 mm in thickness) were arranged alternately and placed between appropriate electrodes. Hence, 10 repeatable cell pairs of diluate and concentrate compartments as well as two electrolyte compartments were assembled. In addition, one CM was used in order to limit the transport of hydroxyl ions generated in the cathode compartment. Before EDBM process, the simulated broth was fed to the diluate compartment, while the sodium alpha-ketoglutarate solution was fed to the concentrate compartment in order to lower the initial resistance on the electrodialysis stack. As an electrolyte solution, 0.3 M sulfuric acid was used and fed to the appropriate electrode compartments. In course of EDBM process at the influence of the applied constant current density, the AKG anions present in the simulated broth migrated through the AM to the concentrate compartment and next they were converted to the acidic form due to H^+ ions generated by the BP. Samples of diluate and concentrate solutions collected at appropriate time intervals were analyzed by HPLC. Each EDBM process was repeated twice and the detailed parameters of the EDBM process as well as the composition of the used EDBM unit are given in Table 2.

In order to determine the efficiency of the investigated EDBM processes, the recovery rate of AKG (*R*), the current efficiency (CE), the specific energy consumption (*E*), and the purity of main product (*P*) was calculated using the following equations:

$$R = \left(1 - \frac{C^t_{\text{dil}}}{C^0_{\text{dill}}}\right) \times 100\% \tag{1}$$

where *R* – recovery rate of AKG, %; C^t_{dil} – concentration of AKG in the diluate solution at the time *t*, g/L; and C^o_{dil} – initial concentration of AKG in the diluate solution, g/L;

$$\text{CE} = \frac{F \cdot z \cdot V \cdot \Delta C_{\text{dill}}}{n \cdot I \cdot \Delta t} \times 100\% \tag{2}$$

Table 1
Main properties of the used membranes

Parameters	AM (PC 200 D)	CM (PC-SK)	BP (PC bip)
Thickness (mm)	0.18–0.22	0.16–0.2	0.2–0.35
Maximum operating temperature (°C)	50	50	50
Transport number	KCl (0.1/0.5N) > 0.91	KCl (0.1/0.5N) > 0.91	–
Resistance (Ω cm)	~2	~2.5	–
Water content (%)	~40	~9	–
Ion exchange capacity (mval/g)	1.24	3	–
Tensile strength (kg/cm^2)	4–5	4–5	–
Water splitting efficiency	–	–	>0.95
Water splitting tension (V)	–	–	1.1–2.2
pH operating range	–	–	0–12

Table 2
The parameters of the EDBM processes as well as composition of the used EDBM unit

Parameters/component of EDBM unit	
Number of membranes	21
Number of cell pairs	10
Active area of each membrane	0.0207 m^2
Thickness of spacers	0.5 mm
Flow rate	100 L/h
Applied current density	90 A/m^{2a}
Electrolyte solution	0.3 M H_2SO_4
Initial concentration of concentrate solution (Na_2AKG)	1 g/L[a]
Initial pH of concentrate solution	8.5[a]
Initial volume of solutions	1.5 L
Temperature of solutions	25°C ± 2°C
Cathode material	Stainless steel
Anode material	Pt/Ir-MMO coated with Ti stretched metal
Pump	Argal, Poland
Source of electricity	DC power supply (NDN)

[a] Established in our previous research [11].

where CE – current efficiency, %; F – Faraday's constant (96,485), C/mol; I – current intensity, A; z – valence of ions; V – diluate volume, L; ΔC_{dil} – change of AKG concentration in the diluate chamber, mol/L; n – number of cell pairs; and Δt – time, s;

$$E = \frac{U \cdot I \cdot t}{m} \tag{3}$$

where E – specific energy consumption needed to produce 1 kg of AKG, kWh/kg; U – voltage, V; I – current intensity, A; m – mass of the final product, g; and t – time, h;

$$P = \frac{C^t_{\text{AKG}}}{C^t_T} \times 100\% \tag{4}$$

where P – purity of the main product, %; C^t_{AKG} – concentration of AKG in the concentrate solution at the time t, g/L; and C^t_T – total concentration of organic acids in the concentrate solution at the time t of EDBM process, g/L.

2.4. Analytical methods

Carboxylic acids and their salts as well as glucose and ethanol present in simulated solutions and samples collected in the time of EDBM process were analyzed using HPLC HP Agilent 1100 Series (Germany). A detailed analytical procedure was discussed in our previous work [11].

3. Results and discussion

The impact of the initial ratio of AKG concentration to those of the other acids present in the simulated solutions, on the efficiency of the EDBM process was investigated. Therefore, two EDBM processes of simulated solutions with different initial concentrations of AKG (15 and 25 g/L, respectively) in diluate solution were compared. Both processes were carried out at the constant current density of 90 A/m^2. In addition, as shown in Fig. 1(a), which illustrates the change in the conductivity of the solution in the diluate chamber at the time of EDBM, the processes were stopped when the conductivity of the diluate solution reached a value below 1 mS/cm. A low value of the conductivity of the diluate solution can be an indication of both, the high recovery rate of AKG ions from the diluate solution and the increase in the concentration of AKG ions in the concentrate chamber, respectively [13]. Moreover, by analyzing the data presented in Fig. 1(a), it can be seen that the initial conductivity of the diluate solution significantly increased at the higher initial concentration of AKG. Low pKa_1 (2.5) and pKa_2 (4.7) values for AKG indicate on a relatively high degree of dissociation of AKG in aqueous solutions in comparison with other acids present in the simulated broths (LA 3.9, AA 4.8, and PA 4.9). On the one hand, the duration of the EDBM process (necessary for lowering the conductivity of the solution in the diluate chamber to 1 mS/cm) increased from 70 to 120 min, when the concentrate solution of higher concentration was used. In addition, as can be seen, a higher amount of AKG was transported through the AM from the diluate to the concentrate

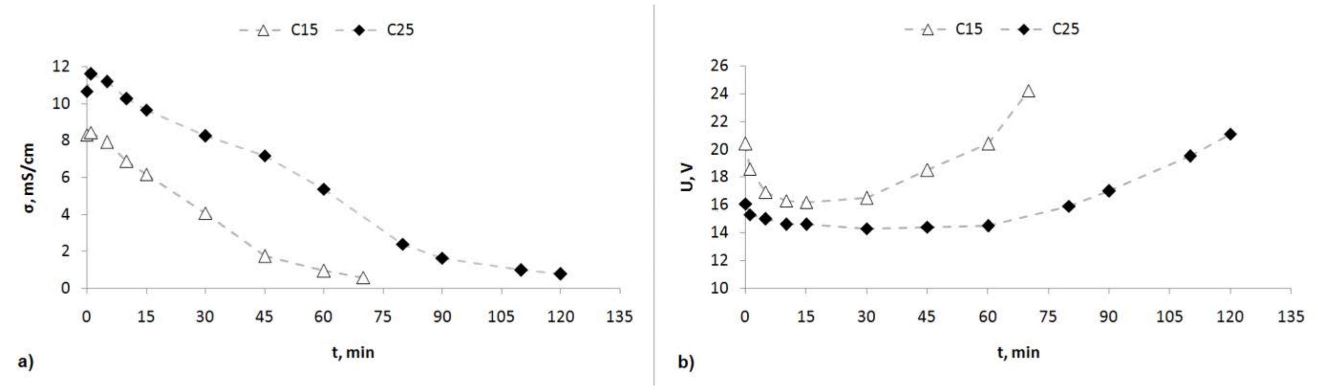

Fig. 1. The change of the conductivity of the solution in the diluate compartment (σ) (a) and the voltage drop on the membrane stack (U) (b) with the time of EDBM processes, $C_{0\text{con}}$ = 1 g/L, i = 90 A/m^2, and T = 25°C ± 2°C.

compartment during the EDBM, when the solution of higher initial concentration was used. Consequently, the increase in the initial conductivity of the diluate solution with increasing AKG concentration led to a decrease in the initial voltage drop on the membrane stack, as shown in Fig. 1(b). It is clear that the increase in conductivity of diluate solution is associated with the decrease of the electrical resistance [14]. Moreover, as reported by Chandra et al. [15], the membrane resistance can be proportional to the concentration of electrolytes and the size of ionic molecules. In addition, in both cases, a decrease in the voltage drop at the beginning of EDBM process can be observed. This effect can be explained by the migration of ions through the AM from the diluate to the concentrate chamber, which leads to an increase in conductivity of the concentrate solution [16]. Furthermore, during the EDBM, the voltage drop stabilizes and then increases until the end of the process, as a consequence of the depleting of AKG ions in diluate solution [17].

Fig. 2 illustrates the changes in AKG concentration in the diluate compartment during the EDBM process. These results indicate that the concentration of AKG in the diluate chamber varied proportionally to the duration of both EDBM processes. Furthermore, AKG was completely removed after 45 and 90 min of EDBM process, when the initial concentration of AKG in the diluate solution was equal to 15 and 25 g/L, respectively. Moreover, for both processes the optimal duration, in which the recovery rate of AKG was 70%–100%, could be specified. Therefore, the continuation of the EDBM process over this optimum time seemed to be unjustified.

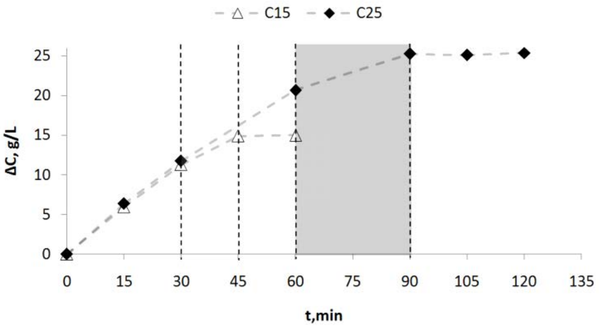

Fig. 2. The change of AKG concentration (ΔC) in the diluate compartment with the time of EDBM processes, $C_{0\mathrm{con}}$ = 1 g/L, i = 90 A/m^2, and T = 25°C ± 2°C.

The obtained results indicate clear differences in the transport of individual acids through the AM, depending on the initial concentration of AKG. Figs. 3(a) and (b) show changes in concentrations of all other acids (LA, AA, and PA) named as by-products, present in the diluate chamber, during the EDBM processes. In the experiment in which the solution with the higher initial concentration of AKG was used, changes in concentrations of LA (4.8 g/L), AA (1 g/L), and PA (0.3 g/L) in the diluate compartment at the end of the determined optimal process duration were smaller. For comparison, when the diluate solution with 15 g/L of AKG was used, the changes in the concentrations of LA, AA, and PA were higher and equal to 7.7, 2.8, and 0.9 g/L, respectively. Two main factors may affect the limitation of transport of by-products with increasing AKG concentration in the diluate solution. On the one hand, the larger amount of AKG in the diluate solution leads to a decrease in its initial pH value. Thus, the by-products which are present in the more acidic solution broth appear mostly in their nondissociated form and thus their electromigration transport is significantly reduced. On the other hand, the greater the amount of AKG ions in the diluate solution is, the greater is the strength of ionic interactions between AKG anions and fixed membrane charges, which can affect the permeation selectivity [15]. In addition, as shown in Fig. 3, the concentrations of individual acids in the diluate compartment decrease in the range: LA > AA > PA according to their initial concentration in the diluate solution and inversely proportionally to their pKa values [15,18,19].

As a consequence of limited transport of the by-products as well as increasing amount of AKG ions transported through the AM, a significant increase in AKG purity could also be observed. As shown in Fig. 4, when the initial concentration of AKG in diluate solution was equal to 25 g/L, the purity (P) of AKG was high for the longer time of the EDBM process and achieved more than 80% at the end of the optimal duration of EDBM process. Moreover, a rapid decrease in the purity of AKG could be seen after 90 min of EDBM process due to an increase in the transport of by-products through the AM after complete removal of AKG ions from the diluate

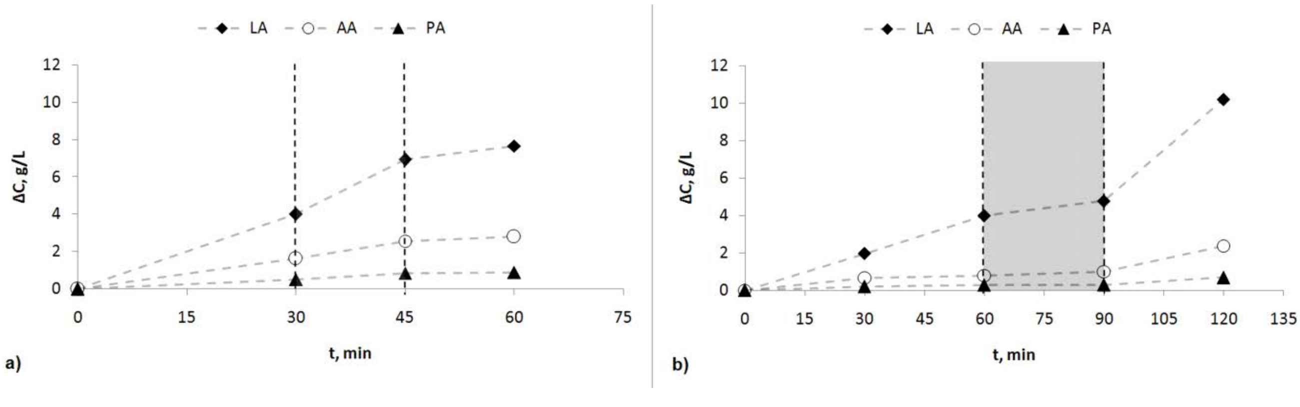

Fig. 3. The change of concentrations (ΔC) of by-products in diluate solution for different initial concentrations of AKG equal to 15 g/L (a) and 25 g/L (b) with the time of EDBM processes, $C_{0\mathrm{con}}$ = 1g/L, i = 90 A/m^2, and T = 25°C ± 2°C.

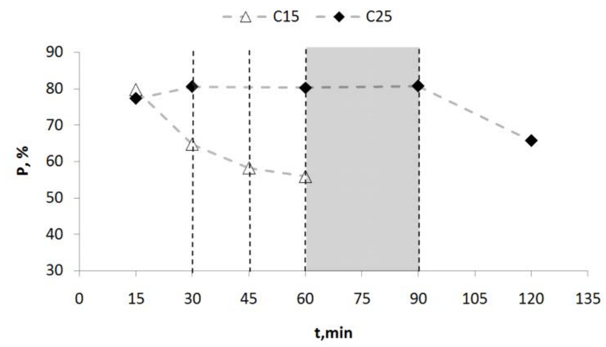

Fig. 4. The change of the purity (P) of AKG in the concentrate compartment with the time of EDBM processes, C_{0con} = 1 g/L, i = 90 A/m^2, and T = 25°C ± 2°C.

solution. For comparison, when the diluate solution with the concentration of AKG equal to 15 g/L was used, the purity of AKG decreased rapidly from 79.7% after 15 min to 58.2% after 45 min of the EDBM process. Thus, the use of the high initial concentration of AKG in the diluate solution in order to achieve a high purity of obtained product of EDBM process seems to be advisable.

In order to determine the economy of the EDBM processes, the relationship between the current efficiency (CE) and the specific energy consumption (E) at the time of EDBM process was analyzed (Figs. 5(a) and (b)). According to the results shown in Fig. 5(a), the calculated current efficiency increased at higher initial AKG concentration in the diluate solution. The reason of such effect is a large number of AKG ions transported through the AM, according to the direction of current and concentration gradient, in particular time intervals [20]. However, it can also be seen that the calculated current efficiency decreased with the time of EDBM, which could be due to the following reasons: (1) an increase in electromigration of by-products at the time of EDBM due to an increase of pH value of the diluate solution associated with generation of OH$^-$ ions at the BP; (2) the leakage of OH$^-$ and H$^+$ through the AM toward the appropriate electrodes [21]; and (3) the competition in migration through the AM between AKG anions and other ions present in the diluate solution [20]. Therefore, the current efficiency (51.3%) calculated for 90 min long EDBM process at the use of the diluate solution with 25 g/L of AKG was slightly lower than the value of CE equal to 60.6% estimated for 45 min long EDBM process at the use of the diluate solution with 15 g/L of AKG. The changes in the specific energy consumption at the time of EDBM processes are shown in Fig. 5(b). As one can see, the specific energy consumption is significantly affected by the voltage drop on the membrane stack and by the amount of produced AKG [22]. Of course, the increase in the initial AKG concentration in the diluate solution conducts to the decrease in the initial specific energy consumption due to the decrease in the total stack resistance [23]. Additionally, it should be noted that in both cases the increase in specific energy consumption with the time of EDBM can be observed. According to the data presented in Figs. 1(a) and (b), as a consequence of the depletion of electrical charge carriers in the diluate compartment at the operation time, the electrical resistance increases [20]. However, regardless of the used initial concentration of AKG in the diluate solution, the specific energy consumption was similar and changed in the range of 1.3–1.8 kWh/kg for both optimal process durations. Summarizing, the increase in the concentration of AKG in the diluate solution leads to the increase in the time of the EDBM process and to the simultaneous slight decrease in the current efficiency. On the other hand, a large amount of the product with significantly higher purity at the same energy consumption is obtained.

4. Conclusions

It is obvious that in order to correctly design the separation and concentration of the main components from the post-fermentation broths, preliminary tests should be carried out on the simulated systems. Hence, we decided to establish the effect of the initial concentration of AKG in relation to other acids (LA, AA, and PA) present in the multi-component simulated solutions on the efficiency of the EDBM process as well as on the final purity of the main product of bioconversion. The results of EDBM process of simulated broths with different initial concentrations of AKG indicate on the possibility of complete removal of AKG from the diluate solution at the constant current density (90 A/m^2) in less than 90 min.

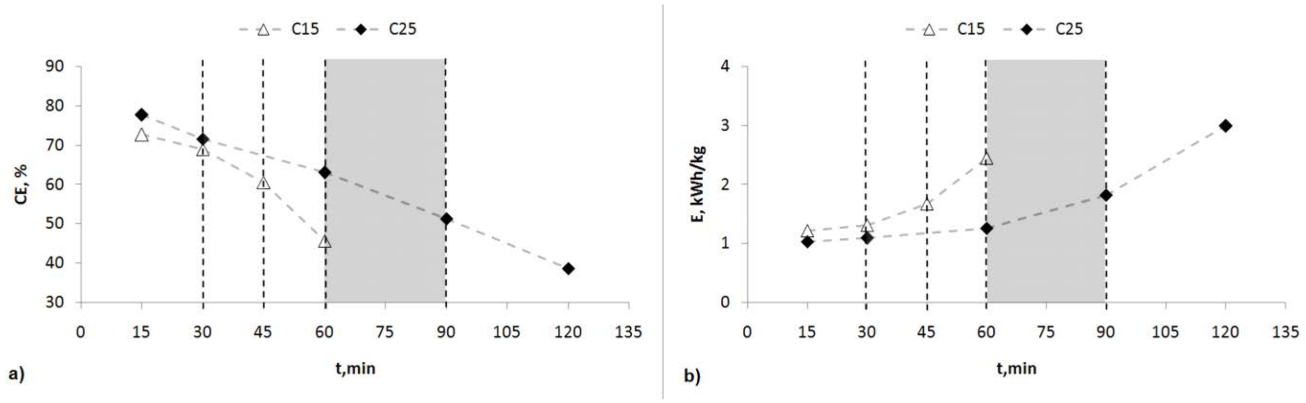

Fig. 5. The change of the current efficiency (CE) (a) and the specific energy consumption (E) (b) with the time of EDBM processes, C_{0con} = 1 g/L, i = 90 A/m^2, and T = 25°C ± 2°C.

Of course, the increase in the initial concentration of AKG in the diluate solution leads to an increase in the duration of the EDBM process (from 45 to 90 min) with the simultaneous slight decrease in the current efficiency (from 60.6% to 51.3%). On the other hand, the increase in the initial amount of AKG in the diluate solution allows for obtaining a much higher purity of the main product (more than 80%) due to the limited transport of by-products through the AM, at the same level of specific energy consumption (1.3–1.8 kWh/kg).

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List of symbols

C^t_{dil} — Concentration of AKG in diluate solution at the time t, g/L
C^o_{dil} — Initial concentration of AKG in diluate solution, g/L
C^t_{AKG} — Concentration of AKG in the concentrate solution at the time t, g/L
C^t_T — Total concentration of organic acids in the concentrate solution at the time t, g/L
CE — Current efficiency, %
E — Specific energy consumption needed to produce 1 kg of AKG, kWh/kg
F — Faraday's constant (96,485), C/mol
I — Current intensity, A
P — Purity of the main product, %
R — Recovery rate of AKG, %
U — Voltage, V
V — Diluate volume, L
m — Mass of the final product, g
n — Number of cell pairs
t — Time, h
z — Valence of ions
ΔC_{dil} — Change of AKG concentration in the diluate chamber, mol/L
Δt — Time, s

Materials

AA — Acetic acid
AKG — Alpha-ketoglutaric acid
EtOH — Ethanol
Gluc — Glucose
LA — Lactic acid
PA — Propionic acid

Membranes

AM — Anion-exchange membrane
BP — Bipolar membrane
CA — Cation-exchange membrane

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