



Circular production – Evaluation of membrane technologies for nutrient recycling from a microbial fermentation effluent

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ABSTRACT

The present study demonstrates a detailed analysis of nutrient recovery from the spent fermentation broth of *S. acidocaldarius*, an archaeon with high industrial and pharmaceutical potential. The pathways of the resource recovery by the lately recognized environmental-friendly and cost-effective technologies nanofiltration (NF), electrodialysis (ED), and electrodialysis with bipolar membranes (EDBM) were assessed. In contrast to previous research, this study treated real wastewater and tested the reuse of recovered products in the subsequent batch fermentations. Ions were separated from the rest of the medium by NF, with 78–85% DOC removal and 0.74 kWh/m³. ED achieved a 1.7 concentration factor for ions, 96% DOC removal and 0.51–0.74 kWh/kg of removed salts. EDBM coupled with NF removed 94.8–98.1% DOC while recovering 0.11–0.15 M sulfuric acid from the feed containing 6.1–16.7 g/l of SO₄²⁻ with energy consumption of 2.7–3 kWh/kgH₂SO₄. Recovered media were successfully applied for control fermentations. While NF was less energy-consuming, ED/EDBM offered a selective ion recovery, higher concentration factors and reuse-specific streams. The discharges of NF/ED/EDBM showed reduced negative environmental impacts due to the nutrient removal from the waste fermentation effluent. All applied technologies require a post-treatment to remove organic substances from side streams and close the production loop with a near-to-zero liquid and waste discharge.

1. Introduction

The growing and aging world population is driving demand for increased industrial development. Among all industrial sectors, the food and pharmaceutical industries have a major impact on excessive water consumption and increased release of toxic substances into land or water reservoirs (Ahmad et al., 2019; Milanese et al., 2020).

Microbial fermentation processes play a substantial part in producing e.g. alcoholic beverages, lactic acid, proteins, and enzymes. Carbon sources, macronutrients (N, P, S), micronutrients (K, Ca, Mg, Na, Fe), and trace elements (Mn, Zn, Co, Mo, Ni, and Cu) dissolved in water (Stanbury et al., 2017) are needed to ensure microbial growth and a high production rate. The practice of excessive supply of nutrients and trace elements to avoid growth and production limitations results in a

nutrient-rich waste fermentation effluent. The spent culture broth may also contain high organic load or toxic substances (Khan et al., 2020; Wei et al., 2021).

Direct discharge of raw wastewater from industrial processes to inland waters, coastal ecosystems, and oceans causes severe damage to biodiversity, eutrophication, ecosystem unbalances, toxic algal bloom and anoxic zones (ESPP, 2021). The destruction of the environment also presents a serious health risk for humans (Porwal et al., 2015). These effluents require wastewater treatment regulation.

The awareness in the food and pharmaceutical industries of the negative environmental effects of effluents and by-products as well as the need for a cleaner and environmentally friendly production is rising (Chandra et al., 2018; Veleva & W. Cue Jr, 2017).

The pollution control emerging from manufacturing industries

Abbreviations: ED, electrodialysis; EDBM, electrodialysis with bipolar membranes; IEM, ion exchange membrane; MF, microfiltration; NF, nanofiltration; SCB, spent culture broth; UF, ultrafiltration; VD, reference medium; CE, % current efficiency; E kWh/kg, energy consumption; Rd %, demineralization rate (total removal efficiency); RE %, removal efficiency; Rs %, recovery/conversion rate; wi, mass fraction.

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themselves has traditionally aimed to reduce the concentration of organic matter and remove potential pathogens in wastewater (Wheatley, 1984). Genetic modifications of microorganisms and recognition of the side effects of e.g. excess phosphorus, sulfate and nitrogen species released in water bodies has led to the introduction of new requirements for wastewater treatment (Howarth, 2008; Wheatley, 1984). The most common treatment methods are wetland, physico-chemical (coagulation/flocculation) and biological (activated sludge) treatment (Ahmad et al., 2019). These methods are mainly focused on wastewater purification without effective waste utilization.

In last few decades, wastewater has become recognized as containing a valuable store of scarce elements such as P and Mg²⁺. Rather than simply regulating nutrient discharge from a regulatory and legislative perspective (European Environment Agency, 2019), novel approaches are needed to synergize nutrient removal and nutrient recycling (ESPP, 2021). Resource recovery is still challenging for full-scale applications (Hamza et al., 2022), but some centralized wastewater treatment plants have recently been retrofitted as water resource recovery facilities (Faragò et al., 2021). Wet chemical technology, crystallization/precipitation and adsorption are commonly applied in nutrient recovery from sludge or sludge-derived ash (Egle et al., 2016; Liu et al., 2021; H. Wu and Vaneeckhaute, 2022). However, end-of-pipe environmental standards should be replaced by pollution prevention through cleaner production, e.g. based on 5 R policy (Olguín et al., 2004). The generation of waste should be tackled at source, comprising reduction, replacement, reuse, recovery and recycling strategies (T. Y. Wu et al., 2009) and diminishing resource scarcity. Membrane technologies, such as nanofiltration (NF), electrodialysis (ED) and electrodialysis with bipolar membranes (EDBM), are seen as attractive methods for nutrient recovery (Blöcher et al., 2012; Wang et al., 2013; Xie et al., 2016; Y. Ye et al., 2020).

NF offers a highly effective means of removing organics and divalent cations. ED is especially promising because it allows removal of organics and concentrates ions and EDBM provides direct acid/base production from ions contained in waste streams (Xie et al., 2016).

The available literature suggests a lack of information derived from experimental research of membrane technologies for nutrient recovery from real wastewater samples. Further challenges include the ability of membranes to retain potentially harmful compounds, which may be present in wastewater but are not included in experiments with synthetic solutions. Moreover, there is a knowledge gap in actual application of recovered nutrients in biotechnological processes. This study offers a path for implementing resource-recovery technologies in the early stage of process planning and optimization to tackle both the resource scarcity and negative environmental impacts of industrial wastewater.

Fermentation with archaeon *Sulfolobus acidocaldarius* is recognized as a future industrial process (Chambers and Patrick, 2015) because archaeal lipids could be used as drug vehicles, vaccine and gene delivery (Benvegnu et al., 2009; Quehenberger et al., 2017). Although the culture medium for *S. acidocaldarius* growth and biomass yield has been optimized via salt and trace elements reduction (Quehenberger et al., 2019), there are still many valuable compounds in the waste fermentation effluent, also known as a spent culture broth (SCB). Sulfuric acid is commonly used for maintaining pH 3 of the substrate during this fermentation, leading to a generation of wastewater with a low pH value and high sulfate concentration. Decentralized SCB treatment could ensure a maximum recovery of the residual nutrients from SCB and their reuse in the same fermentation process, making production and products fit for a climate-neutral, resource-efficient, circular economy (European Union, 2020).

The present proof-of-concept study applied a circular production concept with near-to-zero liquid and waste discharge to recover water and value products from SCB of the biofermentation with the Archaeon *S. acidocaldarius*. The nutrient recovery also contributes to the reduction of pollution potential. Three emerging technologies were investigated in this regard. The indistinct recovery of macro- and micronutrients was evaluated by nanofiltration and ED. EDBM was assessed for the sulfuric acid recovery and base production. Pre-treatment technologies such as micro- and ultrafiltration of SCB were applied to remove mainly organic SCB constituents. Products resulting from the technical recovery processes were reused within an *S. acidocaldarius* fermentation to prove applicability. Microbial growth and production were compared to those obtained by fresh growth medium. The novelty of this study lies in the detailed analysis of nutrient recovery from spent fermentation broth of *S. acidocaldarius*, an archaeon with high industrial and pharmaceutical potential.

2. Material and methods

2.1. Experimental set-up

Fig. 1 shows the process scheme with the technologies assessed in this study. Products from the continuous fermentation with *S. acidocaldarius* were separated from SCB with a ceramic filter. The collected SCB was stored at 4 °C until further use. After the pre-filtration in stage I ("I" in Fig. 1), SCB was further treated by ED (stage II) to increase the ionic concentration or by EDBM (stage III) to produce acid/base. The ED concentrate obtained, enriched with nutrients, was finally used as a cultivating medium in shake-flask fermentation experiments to demonstrate its usability. The acid recovered via EDBM from SCB was reused for the fermentation pH adjustment in the same experiments. The base produced via EDBM may be applied to neutralize the diluents from ED and EDBM. Micro- and nanofiltered SCB were used as an additional control cultivation medium to evaluate the media obtained via ED/EDBM.

2.2. Pre-treatment

SCB (see Supplement) contains organic compounds such as residual microbial cells, peptones, pyruvate, and metabolites that interfere with cell growth (Soma et al., 2021) and must be removed from the recycling process. SCB was microfiltered (MF) after the storage period to ensure removal of suspended solids and proteins. The pH, conductivity and DOC content of the SCB and microfiltered SCB were almost identical. Ultrafiltration (UF) and nanofiltration (NF-DK) were applied in a separate set-up for further removal of dissolved organics and divalent cations of SCB, as EDBM is sensitive to the precipitation of divalent cations on the EDBM membranes (Irfan et al., 2019; Tongwen, 2002). Applied pressures were 4.5 bar, 9 bar, and 20 bar, for the MF, UF, and NF, respectively. Table 1 shows the specifications of applied membranes. The NF270 membrane was used to provide a substrate for the control cultivation.

2.3. ED and EDBM

The experiments were done in a batch laboratory-scale ED system PCCell ED 64-004 (PCCell GmbH, Germany) with ten cell pairs, composed of 9 x PC SA and 10 x PC SK membranes (Table 2). The effective membrane area was 8 x 8 cm². Spacer thickness was 0.45 mm for each stack. Electrodes were Pt/Ir-coated titanium anode and V4A steel cathode, placed in the polypropylene electrode housing material. A

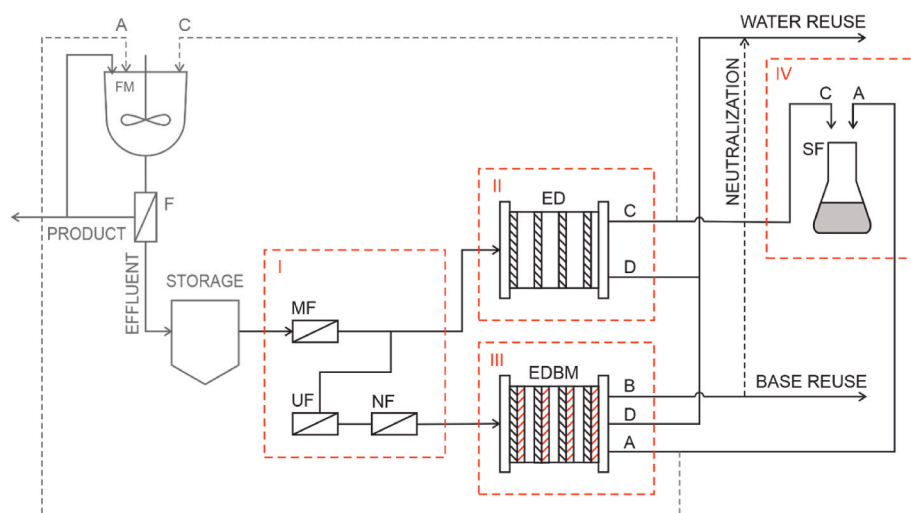


Fig. 1. Process scheme with four assessed stages: I Pre-treatment by MF or UF/NF, II Conventional electro dialysis (ED), III Electro dialysis with bipolar membranes (EDBM), IV Shake flask fermentation (SF). Other involved technologies: FM – fermentation, F – filtration, MF – microfiltration, UF – ultrafiltration, NF – nanofiltration. Relevant streams: Product, Effluent, C – concentrate, D – diluate, A – acid, B – base, Neutralization, Water, and Base Reuse. Processes with grey lines are not in the scope of this work.

Table 1
Membranes applied in the pre-treatment of SCB with the specifications of the manufacturers.

Membrane class	Manufacturer	Model	Material	Stabilized salt rejection (%)	pH 25 °C	MWCO (Da)	Max P (bar)	Max. Temp. °C
MF	SUEZ	JX	PVDF	None	1–11	0.3 μm	5	70
UF	SUEZ	PW	PS/PES	None	1–11	20K	10	70
NF	SUEZ	DK	PA-TFC	98% MgSO ₄	2–10	200	40	80
NF	DUPONT	NF270	PA-TFC	>97% MgSO ₄	2–11	200–400	41	45

Table 2
Ion exchange membranes and their specifications from the manufacturer.

Membrane	Type	Thickness, μm	Transference number	Resistance, Ohm cm ²	Water content (wt%)	pH stability
PC SA	Anion exchange	100–110	>0.95	1.8	14	0–9
PC SK ^a	Cation exchange	100–120	>0.95	2.5	9	0–11
PC MTE	End membrane	220	>0.94	4.5	–	1–13
PC bip ^a	Bipolar	200–350	>0.95 **	–	–	0–12
PC Acid 60 ^a	Anion exchange	100–110	>0.95	2	17	0–9

^a IEMs of the EDBM stack; **water-splitting efficiency.

direct current power supply was applied to adjust the current/voltage driving force.

Current/voltage control was conducted in a stepwise adaptation of the current density based on the decreasing conductivity of the diluate during the ED separation process. Current density-voltage pairs were estimated based on the Cowan and Brown limiting current density method (Cowan and Brown, 1959). More information on current/voltage control is in Supplement. The diluate chamber was fed with 1 L of microfiltered SCB, while the concentrate was filled with 1 L of deionized water spiked with 250 mg/L Na₂SO₄ providing initialization ions for ED. The concentrate chamber was used as a receiving medium for ions from SCB feed, whereas contaminants, proteins, and DOC were mostly retained by the ion exchange membranes (IEM).

Generation of mainly H₂SO₄ acid and bases from the SCB salt content was studied using the same laboratory-scale ED 64-004 PCCell equipment, exchanging the ED stack with an EDBM stack. All IEMs used are described in Table 2 and comprise the same effective area and end

membranes as described above for ED. Diluate was fed with 1 L of nanofiltered SCB (NF-DK), while the acid/base chamber was filled with 0.5 L deionized water. Current/voltage control was conducted by interchanging the constant current/voltage approach. 12 V were applied to induce the separation of water molecules within the bipolar membrane until the conductivity of product solutions reached at least 0.4 mS/cm. Following this, the constant current of 109 A/m² was applied until no significant changes in the electrical conductivity of produced acid/base were observable.

The ED/EDBM feed solutions were circulated at 15 L/h flowrate. Electrode rinsing solution was 0.25 M Na₂SO₄, circulated at 120 L/h flowrate. Process cooling was achieved with tap water circulating through double-wall tanks containing the solutions. Online monitoring of conductivity and temperature was conducted every 2 s. The pH of the feed solution (SCB) was recorded continuously.

Table 3
Overview of shake flask experiments.

Fermentation	Medium	Medium gained through	Acid to set pH
1	VD	Reference Medium	H ₂ SO ₄ (4.8%)
2	ED 1	Electrodialysis	H ₂ SO ₄ (4.8%)
3	ED 2	Electrodialysis	H ₂ SO ₄ (4.8%)
4	NF270	Nanofiltration	H ₂ SO ₄ (4.8%)
5	MF	Microfiltration	Base B2 (EDBM2)
6	VD	Reference Medium	Acid A1 (EDBM1)
7	VD	Reference Medium	Acid A2 (EDBM2)
8	ED 1	Electrodialysis	Acid A2 (EDBM2)

2.4. Analytical methods

Ions in all liquid streams were analyzed by ion chromatography for the following ions: PO₄-P, NH₄-N, anions (Cl⁻, SO₄²⁻) and cations (Na⁺, K⁺, Ca²⁺, Mg²⁺), based on DIN EN ISO 6878, DIN EN ISO 11732, DIN EN ISO 10304-1 and DIN EN ISO 14911, respectively. Trace elements (Co, Cu, Zn, Mo, Cr, and Ni) were below detection limits. Dissolved organic carbon (DOC) was analyzed using the standard DIN EN 1484 method.

2.5. Cultivation of *Sulfolobus acidocaldarius* in recycled media

To minimize evaporation, *S. acidocaldarius* DSM 639 was grown aerobically in a 75 °C temperate shaking oil bath in 100 mL long-neck Erlenmeyer flasks. Shake-flasks were filled with 50 mL medium. The medium described by Quehenberger et al. (2019) was used as a reference medium and is denoted as “VD basal” (= medium without carbon sources). Recovered media used for the growth experiments were from all process steps, including ED1, ED2, NF, and MF gained from SCB, as described in Table 3. Acid A1&-2 and Base B2 are recovered/produced solutions from SCB by EDBM1&-2 (see Section 3.3). The following carbon sources were added to 50 mL of medium (recovered and reference): 1.79 g/L monosodium glutamate (MSG), 3 g/L D-glucose, and 0.5 g/L citric acid. The initial pH was set to 3.0, either with 4.8% H₂SO₄ or recovered acid. All shake-flask fermentations were conducted in duplicate except the reference medium, which was conducted in triplicate. Since the oil bath only holds a limited number of shake flasks, the experiments were performed in two runs. Each run contained a set of three reference medium shake flasks.

2 mL of the culture broth from the shake-flasks were extracted after the lag-phase approximately every 24 h (see Results) to compare the growth of *S. acidocaldarius* in the fermentation experiments using the media described in Table 3. Optical density, OD₆₀₀, was determined with a spectrophotometer (ONDA V-10 PLUS, XS instruments, Italy) at 600 nm against a blank of distilled water. Samples were diluted with deionized water to ensure they remained within the linear range of the photometer. 1 mL of culture broth was centrifuged for 10 min at 10,000 g and 4 °C. The obtained supernatant was used for substrate and metabolite determination by chemical analysis. Glutamic acid and D-glucose concentrations were determined via a photometric assay using a Cedex Bio HT Analyzer (Roche, Switzerland). The concentration of glutamic acid obtained was converted to the used substrate MSG by multiplication by the factor 1.15. Possible metabolites, such as pyroglutamic acid, were identified with an Aminex HPX-87H column (300 × 7.8 mm, Bio-Rad, USA) employing an Ultimate 3000 high-performance liquid chromatography (HPLC) system (Thermo Fisher Scientific, USA). 10 µL samples were analyzed at a flow rate of 0.6 mL/min, and a column temperature of 60 °C. 4 mM H₂SO₄ served as the mobile phase. A RI detector (RefractoMax 520, Thermo Fisher Scientific, USA) and a UV

detector (VH-D10-A, Thermo Fisher Scientific, USA) at 210 nm were used for quantitative determination. The Chromeleon 7.2.6 Chromatography Data System (Thermo Fisher Scientific, Waltham/MA, USA) was used for control and data analysis.

2.6. Data analysis

The slopes of the diluate/concentrate electrical conductivities were calculated to estimate the average rate of total ion migration from the diluate to the concentrate, depending on the ED operation time. Slopes were determined for a specific segment of applied current density as follows:

$$s = \frac{y_n - y_{n-1}}{x_n - x_{n-1}} \quad (1)$$

where x is the ED operation time at two subsequent time points n and $n-1$ on the x -axis, and y are the corresponding diluate/concentrate conductivity values on the y -axis.

Transport phenomena such as osmosis and electro-osmosis were determined from the differences between the slopes (Δs) of the diluate and concentrate for the same ED timeframe and applied current density according to the following equation:

$$\Delta s = s_{\text{diluate}} + s_{\text{concentrate}} \quad (2)$$

where the diluate (s_{diluate}) has a negative slope due to the ionic depletion, and the concentrate ($s_{\text{concentrate}}$) has a positive slope due to the increasing conductivity with increasing ionic concentration.

Total desalination rate (RE), removal efficiency (RE_i), and the ratio of ions present in the ED feed and the ED concentrate (mass fractions w_i) were calculated to assess the ED performance.

Removal efficiency (%):

$$RE = \frac{\lambda_D^0 - \lambda_D^f}{\lambda_D^0} \cdot 100\%$$

or

$$RE_i = \frac{n_i^0 - n_i^f}{n_i^0} \cdot 100\% \quad (3)$$

λ_D [mS/cm] is the conductivity of the feed solution (diluate) at the time 0 and final desalination point; n_i^0 and n_i^f are the molar concentrations [mol/L] of the ion i at the time 0 and the final time, respectively. RE_i in EDBM was calculated based on the number of milliequivalents of anions/cations $n_{Eq,i}$ [mEq] in the initial and final diluate.

Mass fraction:

$$w_i = \frac{m_i^c}{m_i^f} \quad (4)$$

where m_i^c [g] is the mass of ion i transported in each desalination step from diluate to the concentrate solution, m_i^f [g] is the mass of ion i in feed. In the first ED run, the maximum possible value of w_i is 1, meaning that all of the ionic species i is removed from the feed and transferred to the concentrate. The ions were concentrated in the subsequent ED run and the maximum possible value of w_i is 2, denoting the doubled concentration of ion i in the concentrate compared to its content in SCB.

The current efficiency (CE) was calculated to determine how much of the externally applied electrical field is used for the removal of ions from the diluate compartment:

$$CE = \frac{F \sum (n_i^0 - n_i^f)}{N \cdot \int_0^t Idt} \cdot 100\% \quad (5)$$

where n_i [mol] is the mole number of each anion/cation at time 0 and final desalination point. CE is calculated separately either for all cations or all anions, as their removal happens simultaneously but in the opposite directions. F is the Faraday constant (96485 sA/mol). N is the number of cell pairs ($N = 10$ in this case). I [A] is the stack current, and t [s] is the desalination time.

Energy consumption to remove and concentrate ions in the ED set-up and the energy consumption to produce sulfuric acid in the EDBM set-up was calculated:

$$E = \int \frac{UIdt}{c_i V}$$

or

$$E = \int \frac{UIdt}{Mc_i V} \quad (6)$$

where U [A] is the voltage across ED/EDBM stack, c_i [kg/L] is the salt concentration, M [g/mol] is the molar mass of H_2SO_4 ($M = 98.079$ g/mol).

Recovery/conversion rate of salts to acid/base (%) was calculated to determine how efficient the anions/cations are transferred to the corresponding acid/base in the EDBM set-up:

$$R_s = \frac{n_a^f}{n_s^0} \cdot 100\% \quad (7)$$

where n_a^f [mEq] is the final equivalent mole number of acids/bases, and n_s^0 [mEq] is the initial equivalent mole number of anions/cations in feed.

3. Results

3.1. Results of membrane filtration

Filtration steps (Table S2) resulted in 31% DOC removal by UF, 78% by NF(DK) and 89% by NF270. NF(DK) removed above 98% divalent cations, around 30% of the monovalent Na^+ and K^+ , and 40% of SO_4^{2-} . NF270 had lower divalent cation retention (90%) than NF(DK). Thus, the NF(DK) permeate was forwarded to EDBM, whereas the NF270 permeate was directly subjected to the fermentation trials. The energy required for pumps in UF was 0.85 kWh/ m^3 and in NF was 0.74 kWh/ m^3 of SCB.

3.2. Results from ED

Microfiltered SCB was introduced to the batch ED to remove the organic matter and concentrate the ions that were later reused as a substrate for the fermentation. The concentrate/diluate quality and the ED process characteristics are analyzed in the following subsections.

3.2.1. Salt and water transport

Desalination of SCB was analyzed in ED1 to understand the behavior of concentrate/diluate, ion migration, water transport and retention of organic compounds. The desalination was aborted after 103 min, when the concentrate volume increased by 59 ml from the initial 1000 ml, and the total desalination rate was 85%. The ion migration from the diluate to the concentrate was monitored by conductivity measurements for each regime of applied current density (Fig. 2a). The differences (Δm)

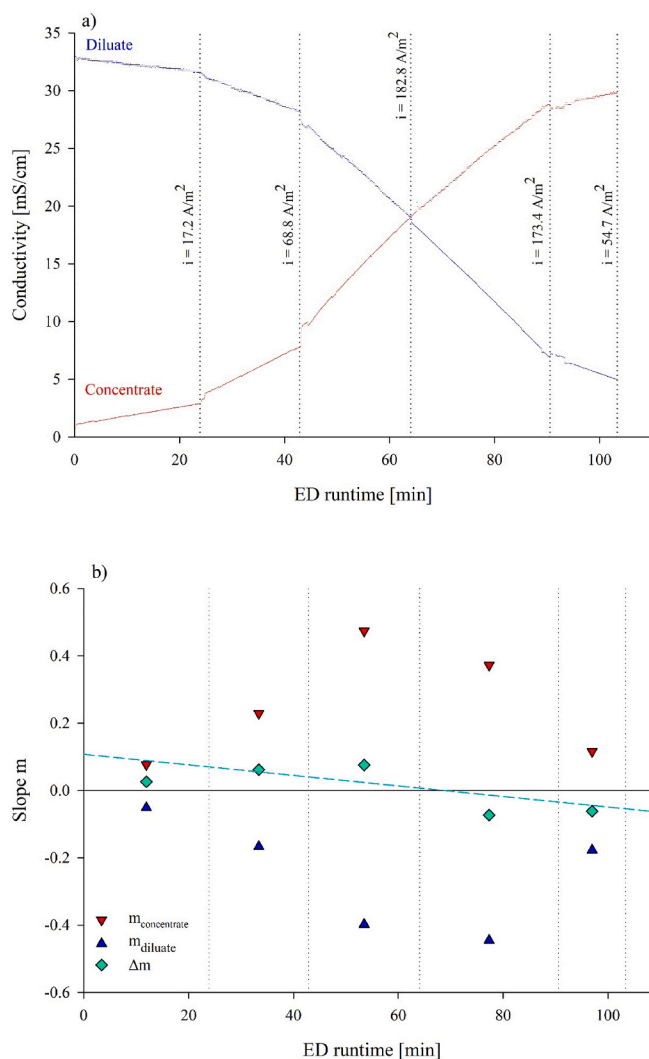


Fig. 2. ED1 desalination process with a) conductivity change in the diluate (SCB) and concentrate over time. Denoted segments correspond to different current densities applied in consecutive steps (dotted line); b) desalination/concentration slopes and their differences for each desalination segment.

between the desalination and concentration slopes indicate the osmotic and electro-osmotic water transport (Fig. 2b). The osmosis and electro-osmosis acted in opposite directions before the conductivities equalized ($\Delta m > 0$) and in the same direction after ($\Delta m < 0$), intensifying water transport from SCB to the concentrate and leading to the limiting concentrate concentration. Besides diluting the concentrate, the water diffusion increased the transport of uncharged pollutants (organic matter and proteins) from SCB that contaminated the concentrate. The DOC measured in the final concentrate was 68.7 mg/l. Since the initial DOC in SCB was 1735 mg/l, the ED thus achieved 96% DOC removal. Energy consumption for SCB desalination was 0.51 kWh/kg of total removed ions (12.5 kWh/ m^3 of SCB).

3.2.2. Concentrating salts from SCB

Doubling the ion concentration from SCB was aimed for in further ED experiments because the microorganisms consume up to 27% of

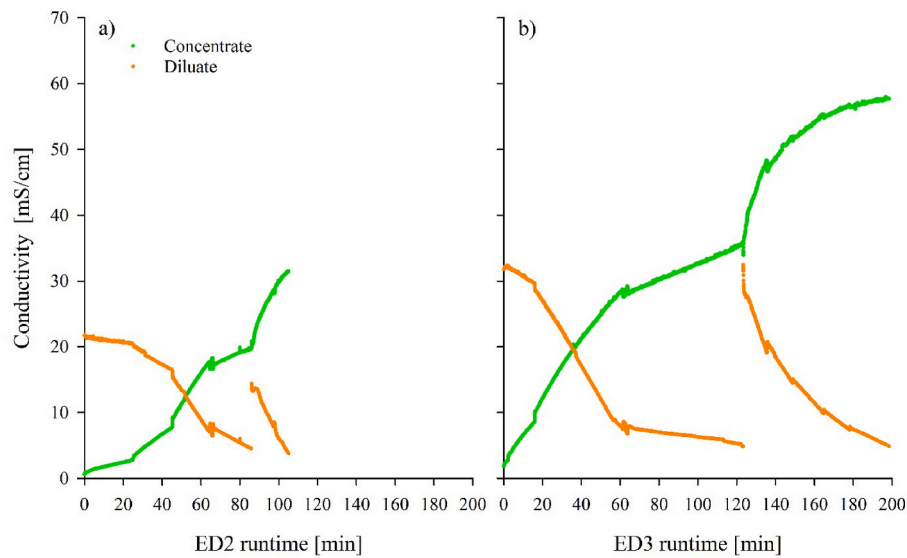


Fig. 3. Doubling the ionic content of the receiving medium from two different feed solutions: a) with lower initial ionic content (ED2 = ED2₁+ED2₂) and b) with higher ionic content (ED3 = ED3₁+ED3₂).

Table 4
Process characteristics of ED2 and ED3.

ED run	Process time	Volume increase in concentrate	RE	CE	Energy consumption
	<i>min</i>	<i>ml</i>	%	%	<i>kWh/kg^a</i>
ED2	105	100	78.6	85.6	0.72
ED3	198	30	84.9	78.1	0.74

^a kg of removed salts.

macronutrient P and 58% of micronutrients (K⁺, Ca²⁺, Mg²⁺) from the reference culture medium (Supplement, Table S1). Concentrating the remaining salts from SCB was necessary to provide the ideal conditions for a new fermentation. Fig. 3 compares concentrating of a lower (ED2) and higher (ED3) concentrated feed (SCB) because waste fermentation effluents often vary in composition. Table 4 summarizes the desalination characteristics. The ions were firstly transferred (ED2₁, ED3₁) and then concentrated in the subsequent batch (ED2₂, ED3₂) by addition of 1 L of SCB to the diluate while retaining the concentrate from the first batch. In both ED2 and ED3, the concentrate conductivity increased ~1.7 times compared to the initial feed conductivity (Fig. 3).

Table 5 presents the ionic mass fractions in each ED run for each ion observed separately as a whole part in feed (SCB). There was a higher fraction of sulfates from SCB (~0.9) and a lower fraction of phosphorus (~0.5) in the first and in the concentrating batch. There was almost no increase of Ca²⁺ and Mg²⁺ fractions when ED3₁ and ED3₂ were compared and K⁺ fractions of ED2₁, ED2₂ and ED3₂ solutions exceeded the maximum reference values.

Table 5
Fraction of ions from SCB transferred to the concentrate for each ED run separately. Reference values denote the maximum theoretical values for each stage.

Ionic mass fraction	PO ₄ -P	NH ₄ -N	Cl ⁻	SO ₄ ²⁻	Na ⁺	K ⁺	Ca ²⁺	Mg ²⁺	Reference
ED1	0.52	0.86	0.99	0.91	0.99	0.70	0.99	0.40	X ≤ 1
ED2 ₁	0.43	0.82	0.80	0.91	0.98	1.32	0.69	0.57	X ≤ 1
ED2 ₂	0.84	1.52	1.77	1.73	1.79	2.65	0.81	0.78	1 < X ≤ 2
ED3 ₁	0.54	0.83	0.54	0.92	1.08	0.63	0.63	0.49	X ≤ 1
ED3 ₂	1.05	1.66	1.14	2.08	2.16	2.40	0.58	0.59	1 < X ≤ 2

3.2.3. Removal of ions in the diluate

The ED diluate was surveyed for any patterned behavior of ionic removal in the first ED batch (ED1, ED2₁, and ED3₁) and subsequent second batch (ED2₂ and ED3₂). The total RE was 82.4 ± 3.4% in all batches, with final diluate conductivities of 4.5 ± 0.5 mS/cm. Fig. 4 shows RE_i of the analyzed cations and anions from SCB. Removal of

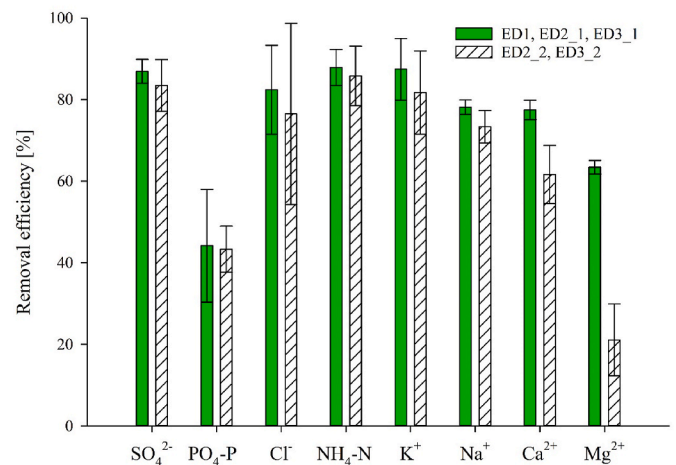


Fig. 4. Removal efficiency of each analyzed ion with the standard deviation bars in the first ED runs (ED1, ED2₁ and ED3₁) for transferring ions, and subsequent ED runs (ED2₂ and ED3₂) for concentrating ions.

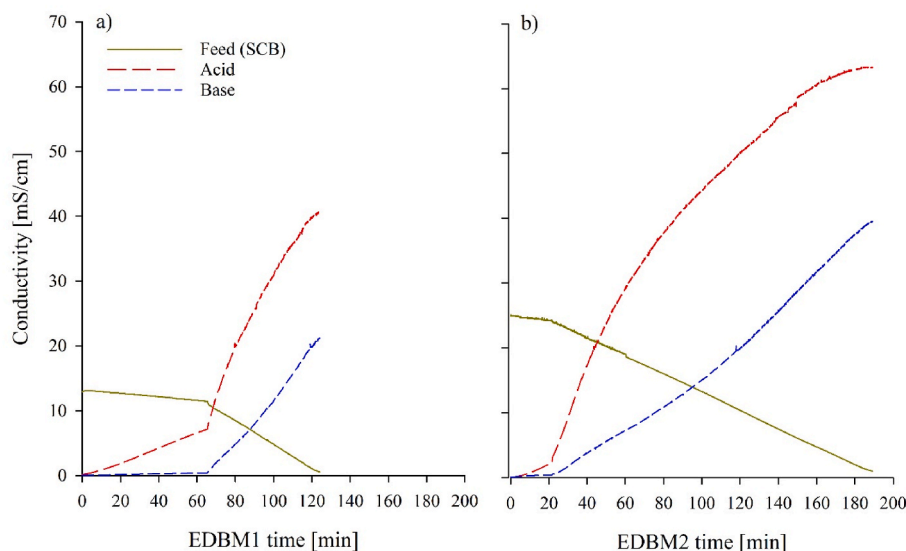


Fig. 5. Conversion of two fermentation effluents with various ionic strengths to corresponding acid/base produced via bipolar membranes in a) EDBM1 and b) EDBM2.

Table 6

Parameters for evaluation of EDBM fed with lower ionic feed concentration (EDBM1) and higher ionic feed concentration (EDBM2).

Parameter	Unit	EDBM1	EDBM2
SO ₄ ²⁻ in feed	mg/L	6512.4	12,172.6
pH acid		1.7	1.5
Anions in feed	mEq/L	133.2	360.9
H ₂ SO ₄ concentration	mol/L	0.11	0.15
DOC in acid	mg/L	8.3	7.5
R _s theoretical	%	52.0	36.3
R _s ^a	%	42.0	22.5
pH base		12.4	12.6
Cations in feed	mEq/L	108.5	280.5
NaOH concentration	mol/L	0.12	0.22
DOC in base	mg/L	13.8	21.6
R _s theoretical	%	54.1	43.4
R _s ^a	%	52.6	41.4
RE	%	95.8	96.1
CE	%	43.2	48.3
Energy Consumption	kWh/kgH ₂ SO ₄	2.7	3.0

^a included intrusion of co-ions, that decreases the actual acid/base concentration.

phosphates was the lowest and remained lower in the subsequent batch. The removal of Ca²⁺ decreased from ~80% to ~60% and of Mg²⁺ from ~60% to ~20%. The concentration of all ions in the diluate/concentrate and the applied current densities are in Supplement, Table S2.

3.3. Results from EDBM

EDBM1 treated the higher concentrated SCB (13.1 mS/cm, pH 3) and EDBM2 treated the lower concentrated SCB (25.1 mS/cm, pH 2.8) (Fig. 5). EDBM2 desalination time was prolonged by 65 min, compared to the EDBM1 process duration of 124 min, due to the almost doubled ionic load of EDBM2 feed.

Table 6 compares the EDBM parameters. The almost double

increased feed conductivity led to 1.8 times higher concentration of produced base, whereas the concentration of recovered acid was 1.3 times higher. Increasing desalination time for EDBM2 slightly increased the concentration of organic compounds in the base. The total removal of organics after EDBM was 98.1% for the acid and 94.8% for the base. Recovery rates (R_s) of acids/bases decreased with increased feed concentration. The acid compartment was especially affected by the co-ion intrusion, with the R_s decreasing by 19.2% and 37.9% compared to the theoretical R_s values for EDBM1 and EDBM2, respectively. Both produced bases contained ~44% of NH₄OH and ~50% of NaOH, corresponding to the concentration of mainly NH₄⁺ and Na⁺ cations in SCB. For the same removal efficiency (RE ~96%), CE and energy consumption were slightly higher for the more concentrated feed in EDBM2 than for EDBM1.

Removal of all assessed ions in the EDBM feed (Supplement, Fig. S1) was in the same range as in the conventional ED (Fig. 4).

3.4. Fermentation with *S. Acidocaldarius* in recycled SCB

Fig. 6 shows the results from the first batch fermentation. From SCB recovered media: ED1, ED2, and NF270 were tested for their usability of cultivation *S. acidocaldarius* compared to the standard medium (VD). NF270 medium showed the most similar results to the standard medium regarding biomass growth measured by the OD₆₀₀ (Fig. 6a). *S. acidocaldarius* growth in ED1 and ED2 medium were close to the reference medium and similar in the substrate uptake (Fig. 6b).

With regard to the outcome of the second run with cultures growing on the MF medium, recovered acids, and a mixture of ED2 and pH set with recovered acid A2 can be seen in Fig. 7. Growth on the MF medium could not be achieved. Both recovered acids procured similar results in terms of growth (Fig. 7a) and substrate uptake (Fig. 7b). The recovered acid combined with ED1 medium did not improve the growth (Fig. 7) compared to ED1 with 4.8% sulfuric acid (Fig. 6).

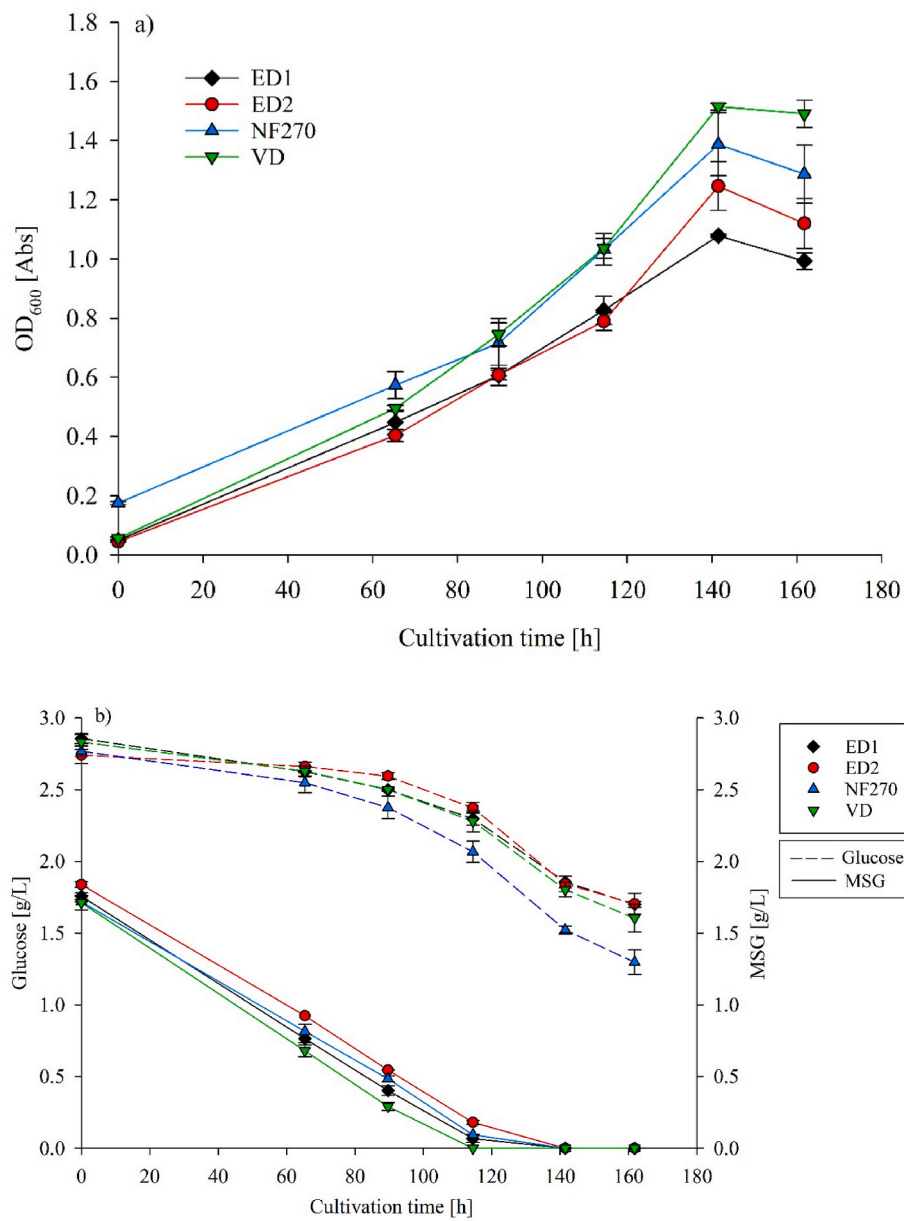


Fig. 6. Results of optical density (OD₆₀₀) measurements in a) and substrate concentration in b) at each sampling point of cultures grown in ED1, ED2 and NF medium compared to fresh medium (VD).

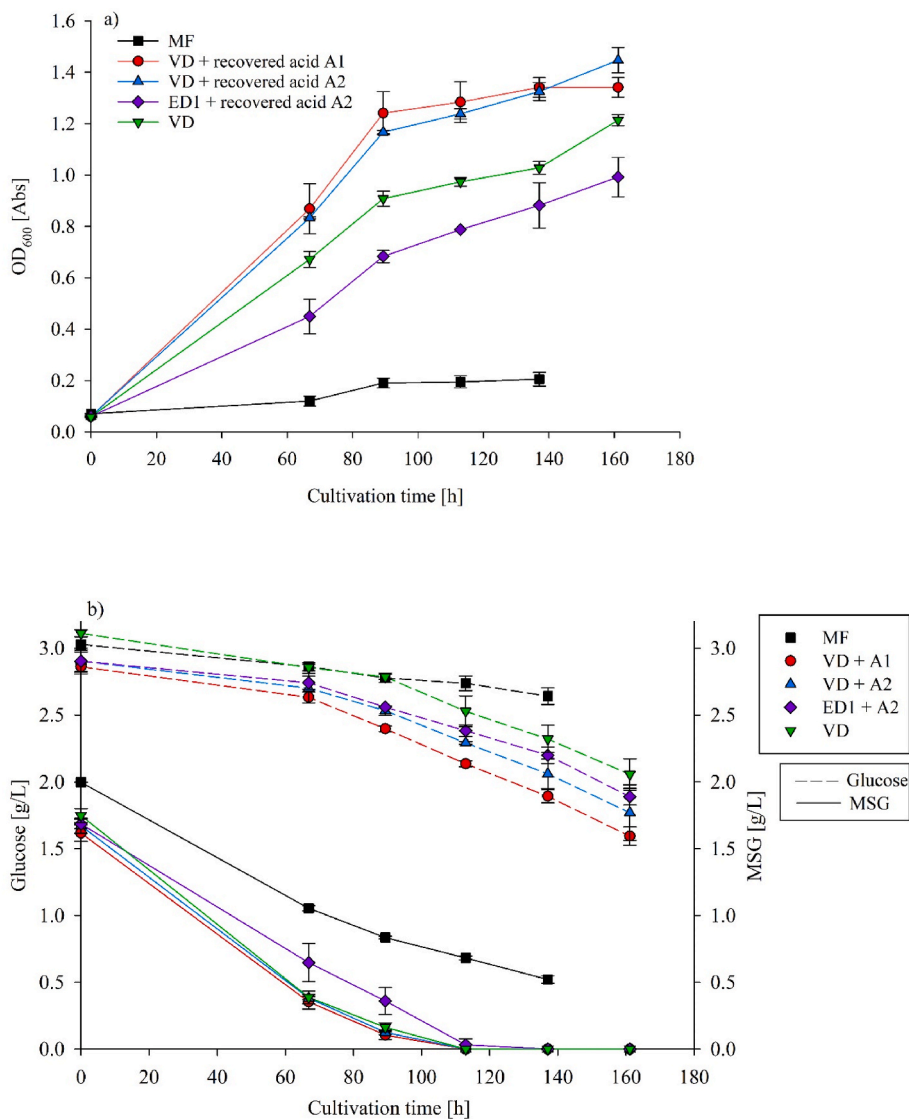


Fig. 7. Results of optical density (OD_{600}) measurements in a) and substrate concentration in b) at each sampling point of cultures grown in MF, VD + recovered acids and mix of ED1 and recovered acid medium. A1 and A2 are the acids recovered by EDBM.

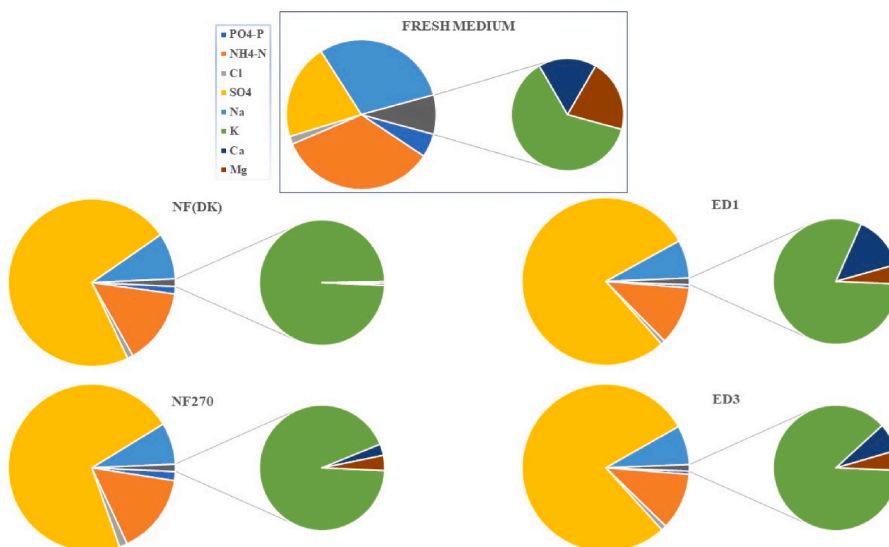


Fig. 8. Contribution of the recovered ions to the total ionic content of recovered media compared to the fresh media composition.

4. Discussion

4.1. Fermentation

Growth on only microfiltered SCB was not achieved. This was probably due to unknown inhibiting metabolites formed in the initial fermentation. Nevertheless, a decrease in MSG concentration over time (Fig. 7b) could be observed. This decrease can be explained by the spontaneous formation of pyroglutamate under high temperatures and low pH (Park et al., 2001). Also, it is possible that MSG was still consumed by the cells, but the uptake did not translate into significant proliferation due to a reduced biomass yield under the inhibiting conditions of the MF medium. Subsequent treatment of SCB via NF or ED can apparently diminish these not yet identified inhibiting compounds. After replenishing the carbon sources MSG and D-glucose, growth on all recovered media, except MF, was possible (Figs. 6a and 7a). The higher recovery rate of SO_4^{2-} in ED compared to NF might slightly decrease the growth curve. Recovered acid by EDMB even showed an improved growth curve (Fig. 7a) as to the reference medium (VD). This effect can be assigned to the impurity of the sulfuric acid and co-ion transport that supply additional P and N sources for microbial growth.

4.2. Pretreatment

Biomass is commonly separated by microfiltration, while fermentation inhibiting substances permeate to SCB (Fig. 7). UF can be excluded from the process (Fig. 1) due to the low DOC removal. NF membranes set off microbial growth and satisfied the EDBM requirements. The permeates had significantly reduced divalent cations and organic foulants, thus decreasing the need for frequent IEM cleaning steps (Merino-Garcia and Velizarov, 2021).

4.3. Nutrient recovery

The composition of macro- and micronutrients recovered by nanofiltration (NF(DK)), single run electro dialysis (ED1) and double run electro dialysis (ED3) from the SCB was compared to the fresh fermentation medium shown in Fig. 8. Pie charts represent only the contribution of individual nutrients to the total amount of nutrients in the medium. Exact values can be found in Table S2.

The ratio of nutrients in recovered media differed strongly compared to the fresh medium due to the sulfuric acid additions and nutrient consumption by microorganisms during the fermentation and losses during the recovery treatment. The microbial growth and production was possible in the media recovered by NF270 and ED (Figs. 6 and 7) as

~40% of P and K sources and ~75% of N sources compared to the fresh medium were available for the microbial growth. This study showed lower P-recovery in ED compared to other ions due to the low pH (3) of SCB wherein phosphorus appears mainly as a monovalent dihydrogen phosphate ion and partially as phosphoric acid (Rotta et al., 2019). The competing transport of other in SCB present ions also decreased the P-removal efficiency (Tran et al., 2015; Zhang et al., 2009). P-recovery rate can be improved by increasing the voltage (Wang et al., 2013), increasing the total effective membrane area (Mohammadi et al., 2021), multistaged ED (Tsiakis and Papageorgiou, 2005), prolonging the desalination time (Tran et al., 2015), or by applying a subsequent ED batch to increase the concentration factor as it was done in this study (ED3 in Fig. 3). The prolonged desalination time would increase the diffusion of nonionic pollutants from SCB towards the concentrate and contaminate it, as shown in Section 3.2.1 and Fig. 2.

ED performed better for micronutrient recovery than nanofiltration with up to 43 times higher Ca^{2+} , 30 times higher Mg^{2+} and 3 times higher K^+ and Na^+ concentrations. Still, divalent cations were significantly less recovered from the SCB (Fig. 4 and Table 5, ED2.2 and ED3.2) due to their lower diffusion coefficients and lower initial concentrations compared to monovalent cations (Długolecki et al., 2010; Feijoo et al., 2021).

The ions initially present in the highest concentration (SO_4^{2-} and Na^+) were effectively concentrated, with a doubled concentration in the receiving medium, followed by other monovalent ions (Table 5). However, all recovered media were unbalanced by the presence of sulfates, which might reach toxic levels to the microorganism in succeeding recovering cycles (Soucek and Kennedy, 2005). Fractionating various nutrient anions and cations from SCB by the application of monovalent selective ion membranes could deliver balanced recovered medium (Z.-L. Ye et al., 2019).

Organic substances were removed up to 96% by ED and up to 89% by NF. Inhibiting substances that were initially present in the SCB were removed by both membrane technologies, allowing the biomass growth (Fig. 6).

4.4. Acid/base recovery

Accumulation of sulfates in the recovered medium (Fig. 8) indicated the alternative applications of EDBM for the sulfuric acid recovery. Bipolar electro dialysis yielded an acidic stream mainly containing sulfuric acid, with around 97% of SO_4^{2-} among all present anionic equivalents. Both bases produced contained 44% of NH_4OH and 50% of NaOH compared to the total concentration of all equivalent cations, corresponding to the high NH_4^+ and Na^+ initial concentrations in the SCB. In

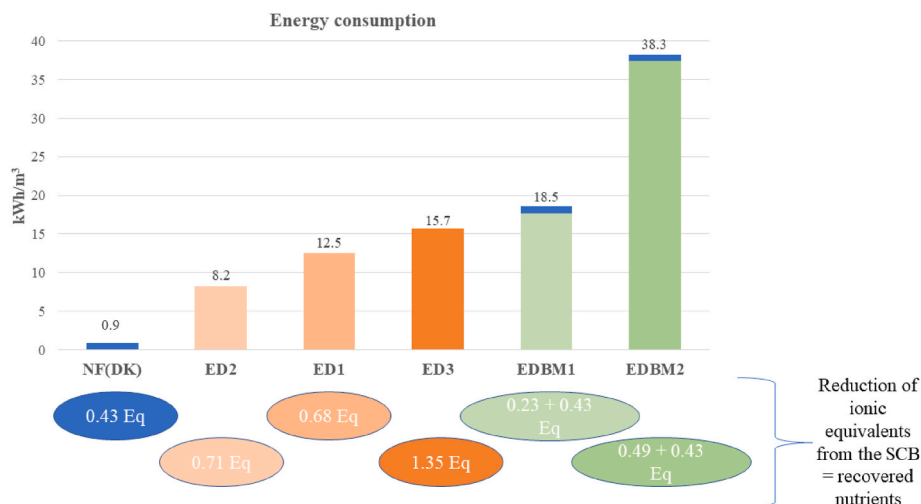


Fig. 9. Energy consumed per m^3 of treated SCB and the ionic equivalents that were removed from the effluents of the each tested technology.

the study by Gao et al. up to 0.9 mol/L H₂SO₄ and 1.6 mol/L NaOH with ~98% purity were recovered by EDBM from the feed with 0.5 mol/L salt concentration (Gao et al., 2021). Concentration of recovered acids/bases in our study were about 10 times lower (Table 6) due to the 10-fold lower initial salt concentration (Table S2), and are similar to those obtained in the study by Marti-Calatayud et al. (Marti-Calatayud et al., 2014) and Lemair et al. (Lemair et al., 2016). Higher concentrated prefiltered feed delivered higher concentrated acid/base (Table 6). The adverse effect of the increasing feed concentrations was the increased co-ion transport, decreasing the final acid/base concentration than theoretically possible (Öner et al., 2021; Tongwen, 2002). Adequate pretreatment with selective increase of the feed salt concentration while decreasing DOC can lead to higher concentrated and purer recovered acid/base, such as ED with monovalent selective anion exchange membranes, as demonstrated in our previous research (Knežević et al., 2022), as well as current density and pH increase of the SCB (Zhang et al., 2012), or increments of volume ratio (Gao et al., 2021).

The pH adjustment with recovered acid was applicable to the batch fermentation, as observed in Fig. 7, but in continuous application it might dilute the substrate and hamper the microbial growth.

The DOC removal was remarkably high for all evolved product streams due to the preceding NF, achieving 98.1% DOC removal for acids and 94.8% DOC removal for bases from the initial SCB organic content.

4.5. Characteristics of discharge streams

A secondary stream evolved after each of the applied membrane technologies for the SCB treatment. The environmental impact and the potential use of discharge streams needs to be taken into account when comparing the applied technologies. NF had a concentrate whereas the ED and EDBM had a diluate stream as the discharge. ED and EDBM significantly decreased the concentrations of nutrients in SCB (Fig. 4 and Fig. S1), preventing severe environmental degradation in case of untreated SCB discharge into receiving water bodies. The greater environmental benefit of e.g. P reduction in the effluent than the resource recovery optimization was presented in the LCA of novel resource-recovery approaches (Fargò et al., 2021). The ionic content of EDBM diluate was lower compared to the conventional ED due to the NF pretreatment (Table S2). The remaining nutrients in NF, ED and EDBM effluents may be suitable for the irrigation purposes if the pH is adjusted by the recovered base obtained in the EDBM experiments. The results of this study also indicate the necessity of a posttreatment application, such as activated carbon or ozonation, for the removal of the inhibiting compounds and DOC reduction.

4.6. Energy requirements

The energy needed for SCB treatment increased with the selectivity of tested technologies NF < ED < EDBM and with the increase of the total ionic content (ED2 < ED1 < ED3; EDBM1 < EDBM2) (Fig. 9). Ionic equivalents removed from SCB have a positive environmental impact due to the waste reduction and their reuse in the fermentation process, resulting in savings for the purchase of new chemicals.

The high energy requirements for ED/EDBM are comparable to the values reported in the literature (Al-Amshawee et al., 2020, p.; Lei et al., 2020; Mohammadi et al., 2021), and most probably offset the benefits of the nutrient recovery, as demonstrated in the study by Fargo et al. (Fargò et al., 2021). Renewable energy to power desalination processes (Al-Karaghoul and Kazmerski, 2013), an increased market for recovered resources, process and membrane optimization can decrease the negative environmental impacts.

5. Conclusions

Our study offers a concept for sustainable production in the future industrial fermentation with *Sulfolobus acidocaldarius*, an archaeon with high pharmaceutical potential. Three technological pathways were contrasted to provide recycled substrate and close the material loop by nutrient and water recovery from the real waste fermentation effluent. Cultivation on the nanofiltered and by ED recovered medium was possible and similar to the reference medium. ED enabled higher concentration factors of recovered macro- and micronutrients with higher DOC removal than NF. Acid recovered by EDBM even exhibited an improved growth curve compared to the reference medium, most probably due to the impurity of the recovered sulfuric acid and co-ion transport that supply additional P and N sources. The recovered nutrients performed well in the batch fermentation, but future studies should seek to integrate the recovery cycle in the continuous fermentation.

NF was economically more feasible than ED/EDBM, but ED/EDBM offered a selective ion separation, higher concentration factors and recovery of high-value products. Further research is needed to reduce the energy consumption of electro-membrane processes and to increase the membrane selectivity in ED for obtaining balanced ion-fractions in the recovered medium and acid/base purity in EDBM.

CRedit authorship contribution statement

Katarina Knežević: Conceptualization, Methodology, Validation, Formal analysis, Investigation, Data curation, Writing – original draft, Visualization. **Kerstin Rastädter:** Methodology, Validation, Formal analysis, Investigation, Data curation, Writing – review & editing. **Julian Quehenberger:** Conceptualization, Methodology, Validation, Data curation, Writing – review & editing. **Oliver Spadiut:** Validation, Writing – review & editing, Supervision. **Jörg Krampe:** Validation, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition. **Norbert Kreuzinger:** Conceptualization, Validation, Resources, Writing – review & editing, Supervision, Project administration, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at <https://doi.org/10.1016/j.jclepro.2022.134436>.

org/10.1016/j.jclepro.2022.134436.

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