Brackish ground water and dairy wastewater treatment using electrodialysis system

Dipak Ankoliya\textsuperscript{1*}, Anurag Mudgal\textsuperscript{2*}, Manish Kumar Sinha\textsuperscript{3}, Vivek Patel\textsuperscript{4}, Jatin Patel\textsuperscript{5}

\textsuperscript{1} Senior Research Fellow, INDIA H\textsubscript{2}O Project, Department of Mechanical Engineering, Pandit Deendayal Energy University, Gandhinagar (Guj.) – 382007, India  
\textsuperscript{2,3} Department of Mechanical Engineering, Pandit Deendayal Energy University, Gandhinagar (Guj.) – 382007, India  
\textsuperscript{3} Department of Chemical Engineering, Pandit Deendayal Energy University, Gandhinagar (Guj.) – 382007, India  
Email: \textsuperscript{2,3} anurag.mudgal@sot.pdpu.ac.in, \textsuperscript{1*} dipak.personal77@gmail.com

Abstract. For brackish water desalination, there is necessity to reduce the volume of saline waste water generated in existing processes due to scarcity of water and to cope up with increasing water demand. The electrodialysis process typically known for its higher water recovery rate with competitive energy consumption per unit volume of clean water produced. Theoretical and experimental investigation of electrodialysis in the feed water composition of Indian ground water would make this technology more adaptable at commercial level. With the capability of bipolar membrane to split water into hydroxide ions and protons, the electrodialysis with bipolar membrane process can produce acid and base solution from the corresponding salt solution. This onsite acid and base production capacity of electrodialysis with bipolar membrane process reduces the need of purchasing these chemicals and also reduces the saline wastewater generation in many industries including dairy industry. This research is aimed to study the electrodialysis system at pilot plant scale installation along with long term testing would make this technology one step ahead in desalination and wastewater treatment.

1. Introduction

Water remains vital for life and crucial common freedoms, for example, livelihood and satisfactory food. Regardless of having covered majority of the Earth’s surface by water, large proportion of population remain inaccessible to the clean water. A portion of the variables adding to developing water shortage are populace development, urbanization, climate change including restricted clean water reserves. It is imperative that we investigate new innovations and take advantage from elective water sources, for example, seawater for our expanding water demands. Despite the fact that seawater is broadly accessible water source, it has high concentration of salts which should be first removed by means of desalination. Desalination is the process to remove dissolved salts and other minerals from the brackish and seawater.

Initially, thermal energy driven processes like multi-effect distillation and multi-stage flash remain market leader for long period of time then very famous membrane technology, reverse osmosis, covers the majority of the desalination market. The growing demand and preference of these pressure intensive membrane processes are due to its simple operation, less space, chemicals and energy requirement and further improvement in materials of membrane [1].
Electrodialysis (ED) is an emerging innovation that can be utilized in the creation of drinking water resource by brackish water desalination. ED can be easily run by the solar energy produced by photovoltaic (PV) panel because ED consumes direct current (DC) for its desalting operation [2]. Due to high recovery ratio, less chemical and energy requirement and high membrane selectivity, ED can become a savvy strategy for the brackish water desalination and wastewater treatment. As an electromembrane process, ED can only allow charged ions to migrate through the special selective membranes under the influence of electric potential and generates two streams one is became rich in ions and other became dilute. ED is widely used in the drinking water production from the brackish water source and table salt production from the seawater [3].

Initial feed water concentration of solution and type of membrane decide the electrical voltage difference required across the ED stack and consequently the energy consumption of the process. The speed at which salt removed from the feed water solution is directly proportional to the electric current. There is also energy requirement to pump the feed water into the ED cell as well as in the electrode compartment. The ED process efficiency strongly depend upon the feed water quality, types of ion exchange membranes and quantity of salt removed from the initial concentration. For any particular feed water, the voltage difference across ED membrane cell vary with the conductivity of membrane and accordingly decides the system efficiency [1]. The use of ED stays restricted by the significant capital expense of system which includes ion exchange membranes, electrodes and replacement cost due to the degradation of membrane polymer.

Electromembrane technologies, for example, electrodialysis and bipolar membrane electrodialysis (BMED) have been generally applied for increasing recovery and valorisation of seawater reverse osmosis reject stream which is called hyper saline stream because of high concentration of NaCl present. The BMED process can covert this NaCl into the acid (HCl) and base (NaOH) by use of electrical energy [4].

BMED is a combination of two processes, one is ordinary electrodialysis and another is splitting water molecule through bipolar membrane. It can make salt transformation and produce H+ and OH− ions without second salt contamination. Due to the economic benefit, technical development and environment friendliness of BMED process, it is considered in vast applications from everywhere in the world, for example, applications in bio food preparation, chemical processes, environmental preservation etc. [5]. As discussed earlier in this section for the ED process, the efficiency of a BMED process is influenced by some of the parameters like fluid flow velocity, current density, cell design, stack layout, spacers and membrane properties.

2. Method
Electromembrane Processes consist of those technologies that apply the electric potential gradient as the driving force of ion transport. They are increasingly important group of separation methods for removal of charged components from solutions such as for producing potable water from brackish water [6].

The main electromembrane processes are:
- Electrodeionization (EDI)
- Electrodialysis (ED)
- Electrodialysis reversal (EDR)
- Electrodialysis with bipolar membrane (BMED)
- Capacitive deionization (CDI)

Lately these processes have started to be applied into vast applications for example energy generation, food processing, personal and health care products, high grade water production, separation of biomolecules and organic materials, sensors, industrial effluent treatment, etc. The basic elements of the Electromembrane technologies can be ion-exchange or bipolar membrane. Ion migration is taking place across these charged membranes which are placed in the electric field.

2.1. Electrodialysis (ED):
In the ED process, when we applied the direct current (DC) voltage across the membrane stack then charged dissolved ions are move through ion exchange membranes. The selectivity of ED process is defined by the electrical charge of feed water contamination. This process allows charged dissolved solids via its ion exchange membrane under the influence of electrical potential while non-charged particles may remain untouched in the brackish water. Due to advances of technology in ion exchange membrane properties and material of construction as well as innovation of polarity reversal, ED process is gaining popularity rapidly [7,8]. The schematic diagram for the working of electrodialysis is illustrated in Figure 1.

**Figure 1.** Schematic diagram of electrodialysis system.

Brackish water desalination for inland water has its own limitation to create it as water resource for example, the amount of concentrate stream water with more salinity than feed water coming from the desalination process would not be a cost effective and environment friendly solution if disposed inland. The higher recovery brackish water desalination system is required to reduce waste management and cost of disposal [9–11].

Electrodialysis process can achieve low volume and high concentration of brine by targeting high (up to 95%) recovery rates [12]. To achieve high water recovery, it is necessary to control the precipitation of sparingly soluble salt like CaSO₄. It is investigated that electrodialysis reversal can achieve higher tolerable CaSO₄ saturation level compared to the RO process due to system’s electrochemical behavior [13,14].

2.1.1. **Design.** Electrodialysis process has following important governing input variables:

- \( C^{fd} \)  Concentration at inlet of dilute stream
- \( C^{d} \) Concentration at exit of dilute stream
- \( R \) Recovery ratio
- \( \Delta \) Thickness of cell pair
- \( u \) Linear flow velocity
- \( Q_{d} \) Product water flow rate (Production capacity)
As all these process parameters are interrelated between each other, it is necessary to choose the chronology between these design parameters and fixing some inputs to calculate all design parameters. Based on these input variables, the aim is to find out length, membrane area, current, voltage and pressure drop across ED stack. By knowing feed water salinity and setting salinity value for the pure output water, we can get salinity value of concentrate stream at outlet by deciding at what recovery ratio ED plant should be run. Here the input salinity of concentrate stream is same as the feed water salinity. By using mass balance equation for both the dilute and concentrate stream, we can find all concentration values [15].

\[ C^c = \frac{C^{fd}R + C^d}{1-R} \] (1)

Here, the \( C^c \) is the concentration at outlet of reject stream, \( C^{fd} \) and \( C^d \) are the concentration of dilute stream at inlet and outlet respectively and \( R \) is recovery ratio. Current density applied is limiting parameter in electrodialysis because it can decide the efficiency of the system. Higher current density can lead to the unwanted phenomenon like water splitting and current leakage which can increase the overall energy consumption of the system. So it is necessary to calculate the limiting current density of the given system which cannot be exceeded at any point of operation of the system to reduce energy loss at the same time increase the full potential of the system. The empirical relation is defined by the Hong-Joo Lee et al. [15] for the limiting current density which is dependent on the flow velocity and outlet concentration in dilute compartment and the geometry of stack. They have also derived formula for the calculation of the membrane area requirement. After calculating the current density, membrane area requirement can be calculated by using concentration, volume flow rate and other constants.

\[ i_{prac} = s a C^d u^b \] (2)

\[ A_{prac} = \left[ \ln \left( \frac{C^c}{C^d} \right) \right] \frac{C^d Q^d}{Q^c} \frac{F \Lambda}{C^d} \frac{1}{\Delta} \] (3)

Where, \( s \) is safety factor, \( a \) and \( b \) are constants and \( u \) is flow velocity. The \( \Delta \) is the cell pair thickness, \( \zeta \) is current utilization and \( \Lambda \) is the equivalent conductance of solution. Current flowing in stack is calculated by using the derived values of current density and membrane area. The electrical potential drop across the cell pair can be derived by using current flow and cell pair resistance in stack. Energy requirement in stack is multiplication of current and voltage applied in stack.

\[ I_{st} = \frac{x F Q^d C^d}{\zeta} \] (4)

\[ U_{st} = \frac{\Delta i_{prac}}{\Lambda} \left[ \frac{1}{C^c} + \frac{1}{C^d} + \frac{\Lambda (pA + pC)}{\Delta} \right] \] (5)

\[ E_{des} = \frac{I_{st} \times U_{st}}{1000 \times Q^d \times 3600} \] (6)

The current flow is denoted by \( I_{st} \), the \( U_{st} \) is the electrical potential drop and \( E_{des} \) is the specific energy consumption of the unit.

2.2. Bipolar membrane Electrodialysis (BMED):
Bipolar membrane electrodialysis process is used to produce acids and bases from a respective salt solution by water separation phenomenon occurred in bipolar membrane by applying electrical potential.

The basic working principle and arrangement of BMED process is shown in Figure 2 having bipolar membranes and cation-exchange and anion exchange membranes which is arranged parallel to form single unit cell between two electrodes. This unit cell consists three compartment and this unit cell is repeated multiple times between two electrodes to form a whole BMED stack. Suppose a feed
solution consisting salt is entered in the middle compartment then under influence of electrical potential applied via two electrodes, the cations and anions in this salt solution will move towards the cathode and anode respectively. Now ions will permeate through the cation and anion-exchange membrane and enter into the adjacent compartment in which hydroxide ions and protons are present which is generated by the bipolar membrane. These anions and cations form an acid and a base by combining with the proton and hydroxide ions respectively. The whole process is resulted into the generation of an acid and a base in BMED cell from the corresponding salt solution.

Technical feasibility and the economics of the BMED process is very much depends upon the performance of bipolar membrane which becomes critical component of this process [16].

![Diagram of bipolar membrane electrodialysis stack](image)

**Figure 2.** Working principle of bipolar membrane electrodialysis stack.

To achieve optimum working point in this electrodialysis process, membrane resistance should be minimum to reduce the energy requirement while current density should be maximum to reduce membrane area requirement.

The design of BMED process is depend on the various parameters like current, voltage drop and membrane area requirement as per the model equations given below [16]:

\[
I = \frac{q_{p}^{F} (c_{l} - c_{s})}{\xi} \tag{7}
\]

\[
A = \frac{q_{p}^{F} (c_{l} - c_{s})}{i \xi} \tag{8}
\]

\[
\Delta U_{cell} = i \left[ \Delta \left( \frac{\ln \frac{c_{l}^{f}}{c_{s}^{f}}}{\Lambda_{s} (c_{s}^{f} - c_{s}^{p})} + \frac{\ln \frac{c_{l}^{f}}{c_{a}^{f}}}{\Lambda_{a} (c_{a}^{f} - c_{a}^{p})} + \frac{\ln \frac{c_{l}^{f}}{c_{b}^{f}}}{\Lambda_{b} (c_{b}^{f} - c_{b}^{p})} \right) + r^{tr} + r^{bml} + r^{am} + r^{cm} \right] + 0.814 \tag{9}
\]

Where, the symbols used in above equations are described in the section 3 with the nomenclature of each symbol in Table 4.

2.2.1. Application. As shown in Table 1, many applications of BMED process were identified under the category of pollution control or resource recovery.
The most established application of BMED process in industrial steel plant is to recover the HF and HNO₃ after pickling bath where the waste flow of KF and KNO₃ is came out. Washington Steel plant in Pennsylvania is the first commercial plant started using BMED process in 1987 [17]. Because of difficulty in the extraction of organic acids after fermentation operation, the application of BMED process in this case becomes very promising [18].

Table 1. Application of bipolar membrane electrodialysis system [17].

| Sr No | Type              | Application                                                                 |
|-------|-------------------|-----------------------------------------------------------------------------|
| 1     | Sulphate recovery | Acid recovery in battery production line                                    |
|       |                   | Converting sodium sulphate waste in rayon manufacturing                     |
| 2     | Acid recovery     | Recovery of acid in steel plant                                             |
|       |                   | Acid recovery from spent aluminium pot linings                              |
|       |                   | Fluorosilic acid conversion to HF, SiO₂                                     |
|       |                   | Control of fluoride emission in chemical plant                               |
| 3     | Nitrate recovery  | Ammonium nitrate conversion from the uranium processing                      |
|       |                   | Conversion of KNO₃                                                          |
| 4     | Pulp & Papers     | Recycling of sodium alkali                                                  |
| 5     | Amine recovery    | Recovery of catalyst in Al coating moulds                                   |
| 6     | Flue gas desulfurization | SO₂ recovery in Soxa²™ process               |

2.2.2. Benefit. To generate H⁺ and OH⁻ ions out of the water by routine method, it requires electrolysis process in which H₂ and O₂ gas is produced which consumes about half of the total applied voltage so the electrical energy of the whole process. These days, double layered ion-exchange membranes called bipolar membranes are available for producing H⁺ and OH⁻ ions from the water without generating unwanted gases. This bipolar membrane with conventional cation- and/or anion-exchange membranes is used to form BMED stack which is used to convert salts into their respective acids and bases [17]. It has low running cost as well as low space requirement.

2.2.3. Limitation. A significant hindrance in the vast commercial application of BMED process is thought to be as per the following [19]:

a) Drastic decrease of the perm selectivity of the membrane due to reduced Donnan exclusion at high product concentrations, which resulted into huge salt spillage via membrane into acid and base.

b) Precipitation of sparingly soluble salt like di- or multivalent salt and organic material at high pH alkaline solution.

c) For very high acid and base concentration, the stability of the anion and cation exchange layer of bipolar membrane becomes very poor.

2.3. Dairy wastewater characterisation:
The quality and quantity of effluent produced in the dairy plant are majorly depend on processing plant parameters like design, operating procedure, production method and kind of product being processed. It also depends on the water management and water conservation program being applied. Dairy wastewater might be separated into three significant classes [20]:

i. Water used in the heat exchanger for heating and cooling purpose are generally less polluted and can be reused or discharged safely.

ii. Wastewaters generated by the cleaning process of various dairy plant equipment like containers, vessels, reactor, pipelines, filters, tanks and other related equipment.

iii. Sewage wastewater generated by sanitary use and application.
The characterisation of effluent water of some of the Indian dairies are shown in Table 2. Important parameters like pH, TDS, BOD and COD of the wastewater at the inlet of effluent treatment plant is taken from the data available in open literature.

Table 2. Dairy wastewater characterisation.

| Sr. No | Parameter                  | Location (city/ state) | Sample collection Source                          | pH  | TDS (ppm) | TSS (ppm) | COD (ppm) | BOD (ppm) |
|--------|----------------------------|------------------------|---------------------------------------------------|-----|-----------|-----------|-----------|-----------|
| 1      | Mother Dairy [21]          | Delhi                  | Combine waste water                               | 7.5 | 1500      | 700       | 2500      | 1200      |
| 2      | Vikas Dairy [22]           | Jalgaon                | Composite waste water                             | 6.5 | 1803      | 729       | 790       | 530       |
| 3      | Sarvottam Dairy [23]       | Bhavnagar              | ETP inlet                                         | 7.8 | ---       | ---       | 2593.3    | ---       |
| 4      | Amul Dairy [24]            | Anand, Gujarat         | Waste water at common storage equalization tank   | 3.69| 3720      | ---       | 7110      | ---       |
| 5      | Vijaya Dairy [25]          | Hyderabad              | Raw effluent                                      | 7.8 | 1100      | 398       | 880       | 372       |
| 6      | Shivamrut Dairy (Jan) [26] | Vijaynagar, Maharashtra Chenna, Tamilnadu | Dairy waste water (whey sample) | 10.0| 1442 | 252 | 1499 | 649 |
| 7      | Madavaram Dairy [27]       | Chennai, Tamilnadu     | Dairy effluent                                    | 5.15| 1128      | 356       | 631       | 216       |
| 8      | Purabi Dairy [28]          | Guwahati, Assam        | Raw dairy waste water                             | 5.64| 722       | 254       | 1462      | 758       |
| 9      | Dairy at Dehradun [29]     | Dehradun, Dehradun, Dehradun, Uttarakhand | Dairy outlet waste water | 7.31| ---       | 455       | 1676      | 770       |
| 10     | Banas Dairy [30]           | Palanpur, Gujaratt, Lucknow, Uttar Pradesh | Sample collected at collection tank untreated wastewater (influent) | 9.93| 1858 | 1766 | 2049 | 1366 |
| 11     | Lucknow Dairy [31]         | Lucknow, Uttar Pradesh | Untreated wastewater                              | 6.2 | 900       | 75        | ---       | ---       |

Min Value | 3.69 | 722 | 75 | 631 | 216 |
Max Value | 9.93 | 3720 | 1766 | 7110 | 1366 |
Average  | 6.81 | 2221 | 920.5 | 3870.5 | 791 |

3. Expected outcomes:
Based on the mathematical equations (1) to (6), the sample calculation can be made for the design of medium scale ED plant to produce potable water. Feed water salinity taken as 1100 mg/L and product water quality can be comparable to RO water as 100 mg/L. ED system design calculation is done for water capacity of 500 LPH with 70% recovery. The calculated input and output parameters are shown in Table 3.

Table 3. Sample calculation for ground water quality for medium scale ED plant.

| Symbol | Description                             | Value | Unit |
|--------|-----------------------------------------|-------|------|
| C^f_d  | Concentration at inlet of dilute stream | 1100  | mg/L |
| C^d    | Concentration at exit of dilute stream  | 100   | mg/L |
| R      | Recovery ratio                          | 0.70  |      |
| ρA+Pc  | Total area resistance of membrane       | 0.0007| Ω m² |
| Q^d    | Product water flow rate                 | 500   | L/h  |
| w      | Width of cell pair                      | 0.1   | m    |
length of flow path per stack $L_{st}$: 0.42 m
Pressure drop in stack $\Delta P$: 350 kPa
Efficiency of pump $\eta_p$: 0.6

**OUTPUT DATA**

Concentration difference of dilute stream $C^\Delta$: 1000 mg/L
Concentration at exit of concentrate stream $C^c$: 4100 mg/L
Concentration at inlet of concentrate stream $C^{fc}$: 3100 mg/L
Practical limiting current density $i_{prac}$: 7.98 A/m²
Area required for stack $A_{prac}$: 12.19 m²
Current passing through stack $I_{st}$: 6.97 A
Potential(Voltage) drop across stack $U_{st}$: 11.02 V
Power required for desalination $P_{des}$: 76.82 W
Specific energy consumption for desalination $E_{des}$: 0.12 kW h/m³

**INPUT DATA**

Concentration of salt at inlet $C_s^f$: 1 eq/L
Concentration of salt at outlet $C_s^p$: 0.5 eq/L
Concentration of acid at inlet $C_a^f$: 0.5 eq/L
Concentration of acid at outlet $C_a^p$: 1 eq/L
Concentration of base at inlet $C_b^f$: 0.5 eq/L
Concentration of base at outlet $C_b^p$: 1 eq/L
Volume flow rate of product $Q_p$: 2.78E-05 m³/s
Faraday constant $F$: 9.65E+07 A s/keq
Current utilization $\xi$: 0.7
Thickness of individual cell $\Delta$: 1.85E-03 m
Current density $i$: 1000 A/m²
Equivalent conductivity of NaCl $\Lambda_s (\text{NaCl})$: 6.32 S m²/keq
Equivalent conductivity of HCl $\Lambda_s (\text{HCl})$: 21.30 S m²/keq
Equivalent conductivity of NaOH $\Lambda_b (\text{NaOH})$: 12.43 S m²/keq
Area resistance of bipolar membrane $\rho_{bm}=r^b+r^m$: 1.00E-03 Ω m²
Area resistance of anion and cation exchange membrane’s area resistance $r_{a+cm}$: 1.60E-03 Ω m²

Expected results for the acid and base production by using BMED stack can be calculated by using equations (7) to (9). If one molar salt solution given as a feed water and reduced to the half molar salt solution then generated acid and base concentration achieved would be the 0.5 molar strength. The generated output data of current requirement, membrane area and voltage drop are shown in the Table 4.

Table 4. Sample calculation for BMED stack.

| Symbol | Value | Unit |
|--------|-------|------|
| $C_{s}^{f}$ | Concentration of salt at inlet | 1 | eq/L |
| $C_{s}^{p}$ | Concentration of salt at outlet | 0.5 | eq/L |
| $C_{a}^{f}$ | Concentration of acid at inlet | 0.5 | eq/L |
| $C_{a}^{p}$ | Concentration of acid at outlet | 1 | eq/L |
| $C_{b}^{f}$ | Concentration of base at inlet | 0.5 | eq/L |
| $C_{b}^{p}$ | Concentration of base at outlet | 1 | eq/L |
| $Q_{p}$ | Volume flow rate of product | 2.78E-05 | m³/s |
| $F$ | Faraday constant | 9.65E+07 | A s/keq |
| $\xi$ | Current utilization | 0.7 | |
| $\Delta$ | Thickness of individual cell | 1.85E-03 | m |
| $i$ | Current density | 1000 | A/m² |
| $\Lambda_{s} (\text{NaCl})$ | Equivalent conductivity of NaCl | 6.32 | S m²/keq |
| $\Lambda_{s} (\text{HCl})$ | Equivalent conductivity of HCl | 21.30 | S m²/keq |
| $\Lambda_{b} (\text{NaOH})$ | Equivalent conductivity of NaOH | 12.43 | S m²/keq |
| $\rho_{bm}=r^b+r^m$ | Area resistance of bipolar membrane | 1.00E-03 | Ω m² |
| $r_{a+cm}$ | Area resistance of anion and cation exchange membrane’s area resistance | 1.60E-03 | Ω m² |

| I | Current required for given capacity plant | 1914.68 | A |
9

| A | Membrane area required | 1.91 m² |
| ΔU_cell | Voltage drop across cell unit | 4.15 V |

4. Conclusion:
Looking at the present scenario of desalination industry and the clean water demand, it is clear that the existing water desalination processes are less efficient in terms of amount of clean water generated from the given input water feed. Electrodialysis process has the flexibility to adjust recovery of the system as well as the degree of desalination required. This unique feature of electrodialysis can provide the new dimensions to the existing brackish water desalination in terms of more productivity and low running cost for the lower feed salinity. Expected results from this study is to demonstrate the electrodialysis process as an emerging desalination technique in the brackish ground water desalination particularly for the Gujarat region. The long term testing of proposed electrodialysis process would increase confidence level in wide spread use of the technology. State of the art pilot scale level configuration of the electrodialysis process at academic institution would open up the gate of collaboration and hybridisation of technologies to improve overall desalination performance. Direct utilisation of DC electrical potential in the electrodialysis and diminishing cost of photovoltaic (PV) solar panel technology would open a door to the potential application in the rural and farthest area from the electricity grid connectivity.

In most of the dairy industries in India generating wastewater with total dissolved solids more than 1000 mg/L will lead to the stringent norms of water disposal. Process of BMED could become a desirable solution with generation of acid and base solution which can be also reused at the same place in the clean in place process. This would create circular economy at the same place with aim of reducing waste.

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