ORIGINAL ARTICLE

# Study on the influence of applied voltage and feed concentration on the performance of electrodeionization

**Yohanes Ervan[1], and I Gede Wenten[2]**

## Abstract

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**Electrodeionization (EDI), a membrane process for removing ion from aqueous feed, becomes more popular nowadays. Its ability to produce ultrapure water gives significant contributions to chemical and semiconductor industries. Only few researches have been published to evaluate its performance in producing ultrapure water, especially in studying and analysing the relation between process parameter and its performance. This work is intended to observe and evaluate the influence of voltage applied and feed concentration on EDI performance. The raw material used in experiment is tap water filtered by reverse osmosis unit before proceeding to the electrodeionization. Product analysis, the diluate as well as the concentrate, is carried out by using conductivity meter and pH meter. The experimental results are presented in i vs V curve, product conductivity vs time, and product pH vs time. From experimental results, it is concluded that concentration polarization can be minimized in EDI process, the increasing of applied current increases product pH. However, there is a limit in gradient concentration between feed and concentrate to minimize the occurence of counter diffusion phenomena.**

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**Key words :** electrodeionization (EDI), ion exchange membrane, ultrapure water

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Nowadays, as the growth of chemical industries increases rapidly, the demand of ultrapure water for industrial process becomes more important. The world market for ultrapure water is currently estimated at around US$ 2.3 billion per annum (Franken T, 1999). The conventional technology for water purification, i.e. electrodialysis is no longer capable of producing ultrapure water, which conductivity reaches 0.055 (S/cm because of the occurence of concentration polarization. Another conventional technology, i.e. ion exchange can produce ultrapure with such quality but the process is not economical because of the cost for chemical reagent to regenerate ion exchange. Since ion exchange process needs periodically regeneration process, hence, process purification cannot be operated continously. As a consequence, we need an alternative technology for producing ultrapure water. Electrodeionization (EDI), a process combining ion exchange and electrodialysis process, is a novel technology for producing ultrapure water that emerges as an alternative solution for producing ultrapurewater with high quality. Besides, EDI can be considered as an environmentally friendly technology.

EDI eliminates the disadvantages of electrodialysis (ED) and ion exchange technology. The electrodeionization cell consists of ion exchange membranes (anion and cation exchange membrane), ion exchange resins (anion and cation exchange resins), a pair of electrodes (anode and cathode) with direct current (DC) as a driving force. Ionic membranes are put in alternating arrangement between anode and cathode, so that many chambers between the two electrodes are created. Aqueous feed containing ionic particles are passed into diluate chambers, which are filled with ion exchange resins. When a current is applied to the electrodes, ions from feed water migrate in different direction according to their electric charges. This migration is controlled by ionic membranes as a barrier for ion migration. The anionic membranes will pass anion and reject cation, as well as cationic membranes will pass cation and reject anion, due to Donnan Exclusion phenomena (Mulder, 1996). Therefore, two types of chambers is created, one contains high ionic concentration, called concentrate chamber, and the other contains very low ionic concentration, called diluate chamber. The principle diagram is shown in Figure 1.

Purification process in EDI contains two main principles that comprised into two steps (Ganzi, G.C., *et al.*, 1992). The first step is a process of which the ions in the feed water are bound by the ion exchange resins, wherein the cations are exchanged with $H^+$ ions and anions are exchanged with $OH^-$ ions. The second step is a process of which the ions bound to the resins are transported by the electrical force through the ion exchange membrane surfaces into the concentrate chambers. These two main principles only occur at the early stage. After the process reaches its steady state, ion exchange resins mainly function as ionic bridge that will increase the overall conductivity of the diluate chambers. Therefore, in EDI process the ion exchange resins hold the key role in exchanging the ions as ionic bridge. Because of the resins existence, the concentration polarization phenomena, which are common in electrodialysis process, can be minimized. Concentration polarization phenomena occur because of the difference in ionic migration rate from aqueous bulk solution to ionic membrane's interface and through membrane's interface (Mulder, 1996). When it functions as ionic bridge, ion exchange in the salt form (after exchanging ion from feed water) will not increase solution conductivity in diluate chamber higher than in its ionic form (in $H^+$ and $OH^-$ form) because conductivity of $H^+$ and $OH^-$ ions is always higher than ion exchange in the salt form. Therefore, regeneration process is needed. In the electrodeionization process, the regeneration process can be done continually without any addition of chemical reagents. The regeneration process occurs due to the $H^+$ and $OH^-$ ions presenting in the solution. The ions are the result of water splitting phenomena, which occur when the ion concentration in the boundary layer is very low due to the occurrence of concentration polarization. Consequently, overall resistance will increase significantly and if this happens

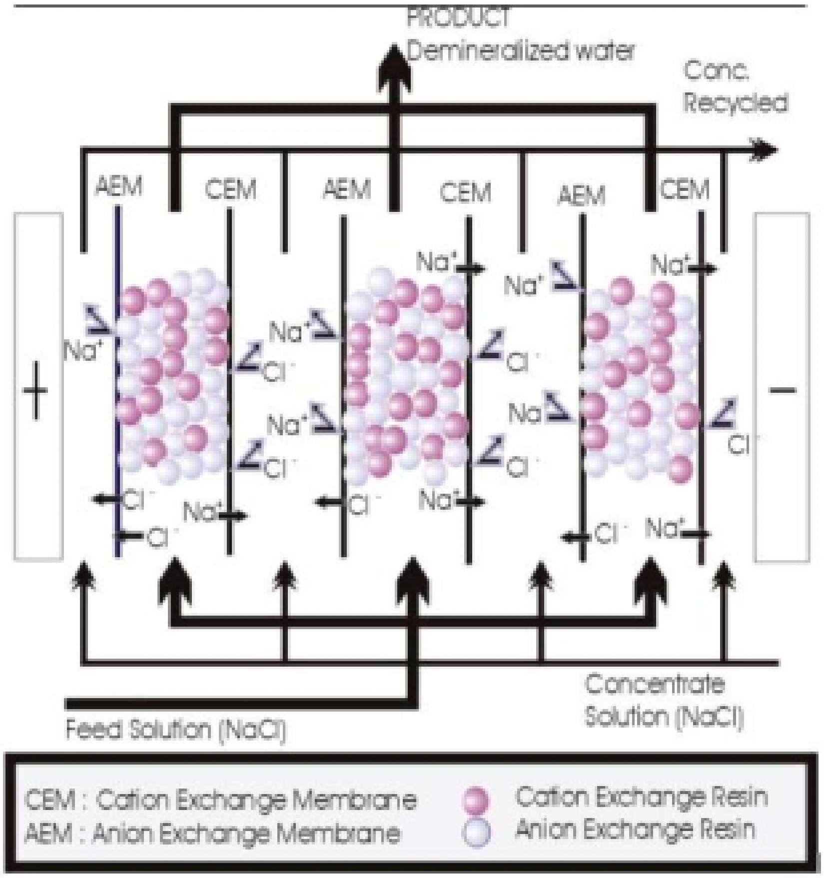

**Figure 1. Electrodeionization process diagram**

continuously then current applied will break water molecule into its ions ($H^+$ and $OH^-$ ion). The water splitting process occurs at the surface of ion exchange membranes as well as at the surface of ion exchange resins.

Only very few experiments have been done in purification process using EDI. Wang J., *et al.* (2000) have successfully demonstrated how EDI process can produce ultrapure water with conductivity reaches 0.05 µS/cm. Spiegel, E. F., *et al.* (1999) uses EDI process for removal low concentration of ammonium ions.

They found out that the using of EDI for removal of low ammonium concentration (<200 ppm) gave an outstanding result, which allowed ammonium concentration in product less than 1 ppm. Few patents have been disclosed related to EDI process apparatus (Sugo, T., *et al.*, 1994; Denocourt, J. P., *et al.*, 1997; Oren, Y., *et al.*, 1998; Nagasubramanian, K., 2000). However, there are many process parameter of EDI that are interesting to investigate and analyze for a better understanding. Therefore, the aim of this research is focused in studying the influence of EDI's process parameter, such as applied voltage and feed concentration, on the EDI's performance.

## Materials and Method

The raw material used in this experiment is tap water which will be processed in reverse osmosis unit before proceeding to the electrodeio-nization process. Schematic experimental diagram is shown in Figure 2. The feed used in elec-

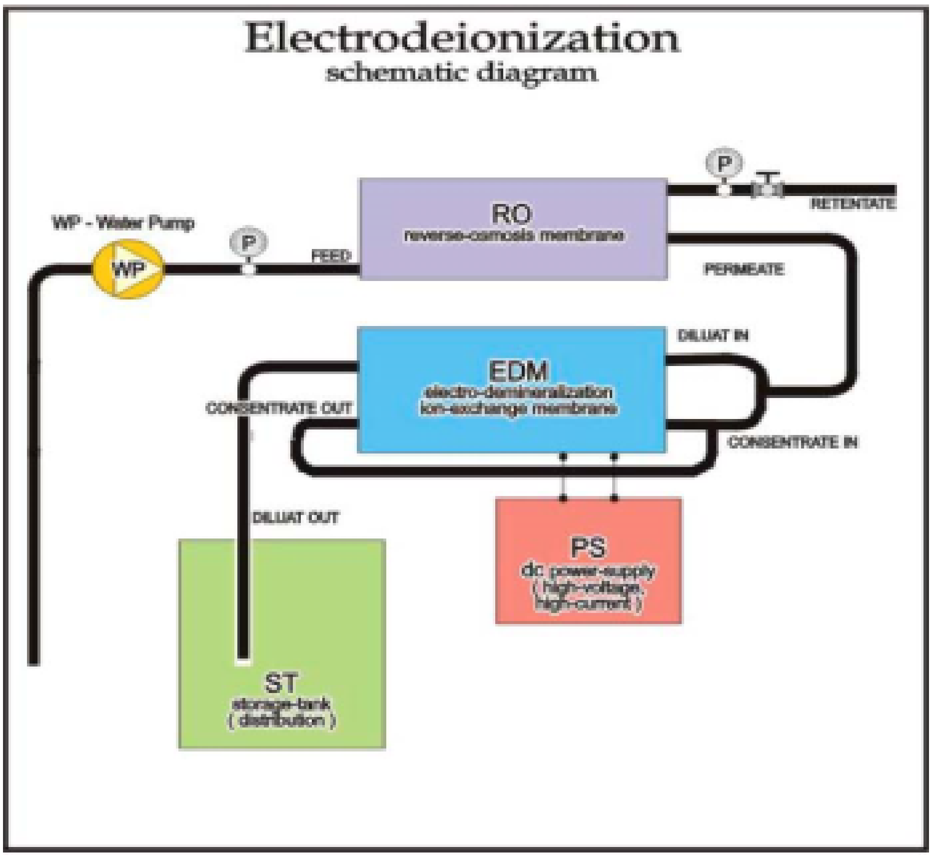

**Figure 2. Experimental Schematic Diagram**

trodeionization unit is permeate of reverse osmosis unit with conductivity reaches 5 µS and the concentrate used is also permeate of reverse osmosis unit with addition of sodium chloride salt to increase its conductivity. The concentrate solution will be recycled back to its storage. The electrodeionization unit has five stacks, consisting of two diluate chambers and three concentrate chambers. The electrodes used are Fe for the anode and Pt for the cathode. Commercially cation exchange membranes MC-3475 and anion exchange membranes MA-3475 are used. The effective membrane area is 0.272 $m^2$. The ion exchange resins are Purolite strong acid cation exchange C-100E and strong base type I anion exchange resins A-400. The resins are placed in the diluate chambers with volume ratio of 30:70 for the cation exchange resins and anion exchange resins, and the width of the spacer is 3 mm.

In this experiment, the varied parameters are current density, feed concentration, and concentrate concentration. The observed variables are product quality in terms of conductivity and pH. The current density variation is carried out by varying electrical voltage applied between 6 V and 50 V. The feed concentration is varied from 5 ppm to 80 ppm, and the concentrate concentration is varied from 30 ppm to 180 ppm.

## Results and Discussion

### i-V curve characteristics

Mulder (1996) reported that during electrodialysis (ED) of salt solution, a zero gradient line region has been observed in current density versus voltage curve. The existence of zero gradient line region means that the overall resistance in diluate chambers increases significantly because of the occurrence of concentration polarization phenomena. As mentioned before that the concentration polarization happens because of the difference in rate of ionic migration. Consequently, at low ionic feed concentration, there are not enough ions in diluate chambers, which can be transported by electrical current so that the overall resistance in diluate chambers increases significantly.

However, when compared with the EDI process, it is not in agreement with what is usually observed in electrodialysis. Figure 3 depicts curves describing the relation between current density and applied voltage resulted from ED and EDI operation. It can be seen from Figure 3 that a zero gradient line region is not shown during ED operation from conducted experiment. Although it cannot be compared perfectly for both of processes, there is still a qualitative difference, which can be observed. EDI's curve shows a tendency of decreasing resistance and there is nozero gradient line region, while in ED the increase in resistance is proportional to the increase in electrical voltage. It means that the concentration polarization can be minimized in EDI process. This result is in agreement with the experiment that had been conducted by Wang, *et al.* (2002). From Figure 3, it can also be noticed that the increasing of applied current follows Ohm's law until 40 Volt. Beyond that voltage, the increasing of applied current is no longer following Ohm's law, but it increases non linear due to the excessive $H^+$ and $OH^-$ ions in diluate chambers.

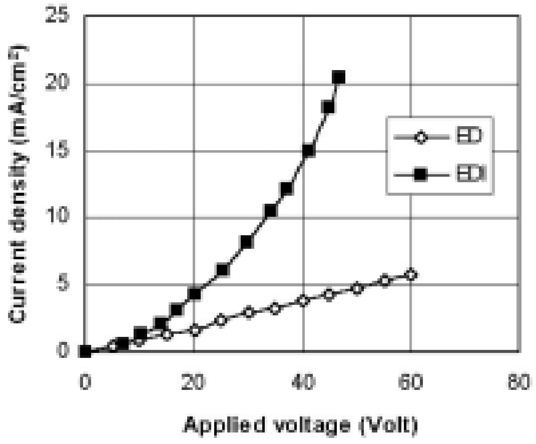

**Figure 3. Current density versus applied voltage curves in EDI and ED processes**

## Current density variation

By varying the electrical voltage until 50 volt, the produced product has a very good quality, which is almost 0 ppm (the exact quality cannot be measured due to the limitation in the instrument used). The experimental results show that the increase of applied current density does not cause significant effect to the product quality although the process has only been operated for about 20 minutes. The amount of ions transported through the ion-exchange membrane is directly proportional to the electrical current density. The increase of the current density leads to an increase in the number of ions transferred. The electrical current required to transport a number of ions is given by:

$$I = \frac{\Delta C \times Q \times F \times \text{valency}}{10^6 \times MW}$$

I (A) = Theoretical current density needed for purification process
C (ppm) = Concentration gradient between feed and diluate
Q (ml/s) = Feed flow rate
F = Faraday's constant as 96500 ampere-seconds/eq
MW = Molecular weight (in this experiment used NaCl with MW as 58.5)

Theoretically, the current density required to process 2.44 ppm feed concentration with volumetric flow rate of 17 ml/s is 0.25 ampere. By applying current density of 4.04 ampere (in 20 V electrical voltage), the current efficiency is only 6%. Current efficiency can be calculated from:

$$\text{Current efficiency } = \frac{\text{I theory}}{\text{I applied}}$$

A-6 percentage current efficiency means that only 6 percent from total applied current is used to transport ions from diluate to concentrate chambers, while the rest is lost as a generated heat and used to break water molecule into its ions. Therefore, the applied electrical current is more than enough to carry out the separation process.

The curves that show concentrate and diluate pH versus time is shown in Figure 4 and the effect of electrical voltage variation to the product and concentrate pH can be seen in Figure 5 and Figure 6, respectively. From Figure 4, it can be observed that there is difference between diluate and concentrate pH at 20 Volt. It can be seen that diluate pH tends to increase, while concentrate pH tends to decrease at the early stage of process. At further operation, concentrate pH will increase until pH 6, and diluate pH will decrease until pH 7. It is concluded that at this applied voltage (20 V), ion exchange resins in diluate chambers still can maintain the neutrality of diluate pH. The decreasing in concentrate pH is resulted by $H^+$ ions transport from diluate chambers. Furthermore, the decreasing in concentrate pH means that water splitting phenomena occur even at low applied voltage.

By increasing the applied current density, the current efficiency becomes lower and the product pH will be increased, as shown in Figure 5 and Figure 6. Theoretically, the ion exchange resins placed in mix bed form in the diluate chamber can maintain the neutral pH of the product. However, an increase in the applied current density leads to a tendency of the increasing in product pH and decreasing of concentrate pH. Iurash, *et al*. (1999) studied the effect of volume resin composition on concentrate and diluate pH and found that larger volume fraction of anion resin will shift solution pH into base. Therefore, while conducted the experiment with ratio anion to cation resin is 70:30, the given result is in agreement with what has been observed during this experiment, i.e. the concentrate pH tends to decrease and diluate pH tends to increase. However, further researches must be done to give a

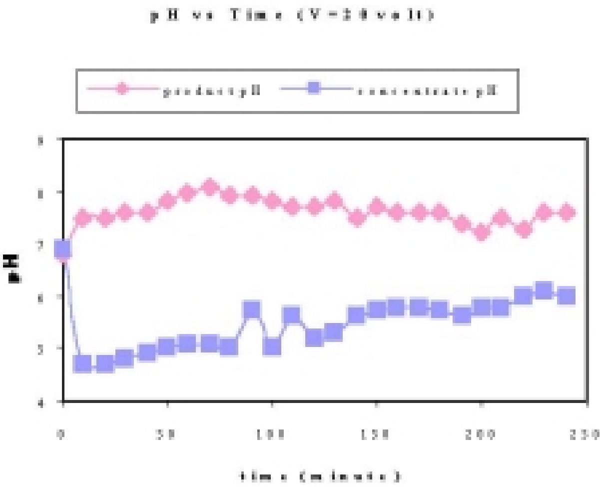

**Figure 4. Concentrate and Diluate pH versus Time curve**

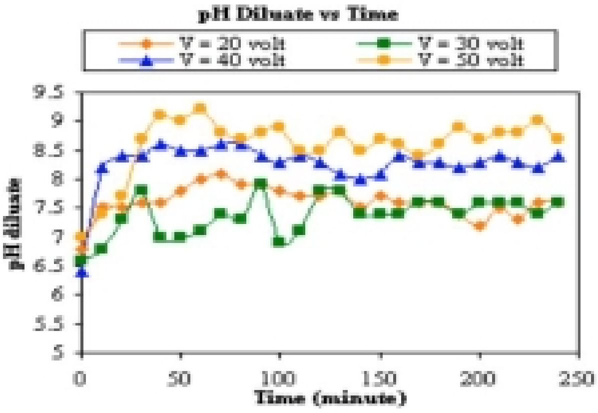

**Figure 5. Diluate pH vs. time**

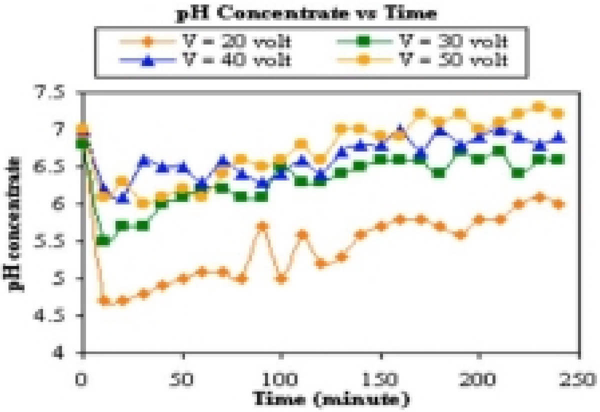

**Figure 6. Concentrate pH vs. time**

better understanding about the relation between volume resin composition and solution pH on multilayer ion-exchange resin since Iurash, *et al.* (1999) have only experimented with monolayer ion-exchange resin placed in diluate chambers. In addition, the pH tendency in both diluate and concentrate chambers is estimated as a result of unevenly spreading of cation and anion exchange resins. Due to this, it is possible that in the water splitting region there are not enough anion exchange resins to transport the anions from the diluate to the concentrate chamber.

If the theoretical applied current density in the demineralization process is known, it can be estimated the current density requirement with high current efficiency. Nevertheless, in the demineralization process, it is common to use less than 100% current efficiency. It is because some fraction of the current applied is required to regenerate the ion exchange resin through water splitting.

## Counter diffusion phenomena during electrodeionization process

In an EDI system, ion exchange membranes are used in combination with an electrical potential difference. Two forces act on the ionic solutes; concentration difference and an electrical potential difference. Under there circumstances, transport of an ion can be describe by a combination of these two processes, i.e. a Fickian diffusion and an ionic conductance. The equation is known as the Nerst-Plank equation (Mulder, 1996):

$$J_i = -D_i \frac{dc_i}{dx} + \frac{z_i F c_i D_i}{RT} \frac{dE}{dx}$$

The diffusion (counter diffusion) process happens in reverse direction compared to transport ions by electrical potential difference. This diffusion will effect the overall process performance since ionic removal by electrical potential will balance by ionic transport from concentrate to diluate chambers. The greater concentration difference in both chambers (concentrate and diluate) the faster ionic migration to diluate chambers and the lower process performance. This phenomenon is due to larger concentration in the concentrate than in the feed chambers and hence reducing overall electricity resistance during process. This counter diffusion will not affect the process performance as long as counter diffusion rate can be balanced by migration rate. However, if counter diffusion rate is larger than the migration rate, contaminant ions from concentrate chamber will reduce product's quality.

In this experiment, the parameter variations conducted is feed and concentrate concentration. The result is presented in curves showing a relation between % separation versus time (Figure 7). It can be seen that both concentrations has significant influence on percent separation. With varying feed concentration at a fixed 180 ppm concentrate concentration, it can be noticed that percent separation increases with feed concentration. However, this tendency is in contrast if concentrate concentration is varied at a fixed 3.2 ppm feed concentration. The increase in concentrate concentration results in the decreasing of percent separation.

While feed concentration is varied from 5 ppm until 80 ppm, the increasing in concentration gradient between feed and concentrate concentration cause a reduction in percent separation. The increasing in gradient concentration between feed and concentrate accelerates the counter diffusion from concentrate to diluate chambers. Counter diffusion rate can be balanced until gradient concentration reaches 170 ppm or less. However, if EDI is operated at gradient concentration larger than 170 ppm, the ionic migration from diluate to concentrate is no longer capable to balance the counter diffusion rate. This can be observed when EDI is operated at 5 ppm feed concentration (gradient concentration is 175 ppm), which gives a poor separation (the value of percent separation is -100%). At this condition, percent separation is poor, implying that product concentration is higher than feed concentration.

The same phenomena also occur when concentrate concentration is varied. The rate of counter diffusion can be balanced until gradient concentration between feed and concentrate is less than 160 ppm. If we operate at 180 ppm con-

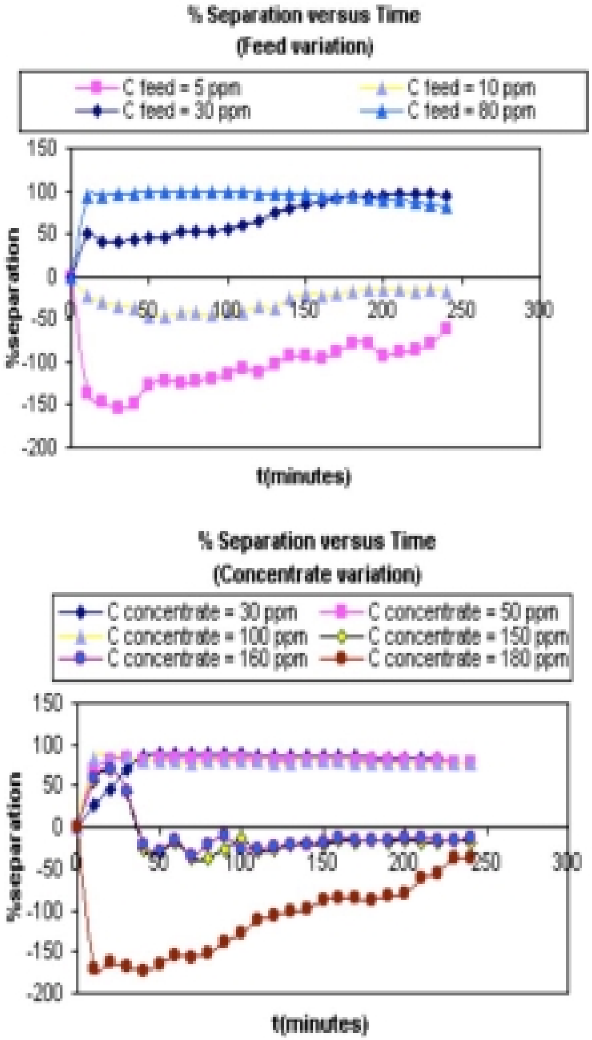

**Figure 7. Concentration variation versus time curve (C feed = 3.2 ppm, C concentrate = 180 ppm).**

centrate concentration (gradient concentration is 177 ppm), poor separation is achieved (the value of percent separation is -150%).

When further examination is made on both curves (Figure 7) percent separation will be found to increase with the increasing time. This happens when EDI is operated at gradient concentration larger than 170 ppm in feed concentration variation or at gradient concentration larger than 160 ppm in concentrate concentration variation. The percent separation increases until it reaches value of -5% separation. The phenomena occur because of the decreasing in concentrate concentration (in this experiment, concentrate is operated in batch system with recycle, thus, concentration concentrate will be decreased because of the dilution process). Consequently, the driving force of counter diffusion will decrease and result in the decreasing of counter diffusion rate. The value of -5% separation is reached when gradient concentration is about 160 ppm on both curves. With this gradient concentration, counter diffusion has the same rate as ionic migration from diluate to concentrate, thus, percent concentration remains in the same value as time increases. It can be concluded that ion in EDI's chambers have reached equilibrium state. Furthermore, it can be concluded that EDI should be operated in gradient concentration lower than 160 ppm in order to minimize the influence of counter diffusion phenomena.

## Conclusions

1. It is shown that purification process by electrodeionization give an outstanding result in product quality, which can reach almost 100 % rejection continuously, without the need of periodical regeneration, as in ion exchange process.
2. The disadvantage of electrodialysis process due to the concentration polarization can be minimized in the demineralization process by electrodeionization method, by placing ion exchange resins in the diluate chambers.
3. The increase of applied current voltage that causes the decrease of current efficiency resulting in the increase of product pH. The neutrality of product pH can be maintained in current efficiency of 6% or more.
4. It is observed that counter diffusion phenomena can be minimized when EDI is operated under concentration difference lower than 160 ppm.

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