Continuous microalgae recovery using electrolysis with polarity exchange

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ABSTRACT

There is increasing interest in the use of microalgae as a renewable source for the production of fuels and chemicals, but improvements are needed in all steps of this process, including harvesting. A continuous microalgae harvest system was developed based on electrolysis, referred to here as a continuous electrolytic microalgae (CEM) harvest system. This innovative system combines cultivation and harvesting and enables continuous and efficient concentration of microalgae. The electrodes were subject to a polarity exchange (PE) in the middle of the operation to further improve the harvest efficiency. Use of PE, rather than conventional electro-coagulation-flotation (ECF), led to more efficient cell recovery and more uniform recovery over the entire harvest chamber. In addition, PE increased the cell growth rate and the circulated cells remained intact after harvesting.

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1. Introduction

Microalgae produce oils through photosynthesis and are widely accepted as the only source of biodiesel that can meet the global demand for renewable and green fuels for transportation (Chisti, 2007; Gouveia and Oliveira, 2009). Microalgae have the potential to produce up to 100-fold more oil per unit biomass than land plants (Chisti, 2007), but mass production of oil from microalgae has technical obstacles that limit development of this industry (Chen et al., 2011). In 2006, the price of palm oil was US$0.52/L, but microalgal biodiesel was US$2.80/L (Chisti, 2007). Thus, it is crucial to reduce the cost of microalgal biodiesel production, including the harvest costs (ca. 20–30% of the total) which include cultivation, harvesting and drying, extraction, and conversion (Mata et al., 2010).

The grand challenge is to efficiently harvest tiny cells from extremely dilute solutions (Vandamme et al., 2011). Thus, many techniques have been proposed for the harvest of microalgae, such as centrifugation (Rodolfi et al., 2003), flocculation (Bilanovic and Shelef, 1988; Kim et al., 2011; Sukenik et al., 1988; Vandamme et al., 2010), filtration (Danquah et al., 2009a,b), flotation (Chen et al., 1998; Uduman et al., 2010), ultrasound (Bosma et al., 2003), pH (pH 11) adjustment (Lee et al., 1998), and electrolysis (Vandamme et al., 2011). With the exception of the electrolytic method, all of these techniques are considered unsuitable because of poor energy efficiency and operational complications, both of which appear difficult to improve (Uduman et al., 2010).

The water industry has long used electrolysis-based technologies for the removal of contaminants, including algae (Alfafara et al., 2002; Boroski et al., 2009; Feng et al., 2003; Ilhan et al., 2008; Meas et al., 2010; Szpyrkowicz, 2005). This approach concentrates microalgae by the following reactions (Gao et al., 2008; Meas et al., 2010; Szpyrkowicz, 2005): (a) aluminum ions are liberated from a sacrificial anode through electrolytic oxidation (Eq. (1)); (b) aluminum ions undergo hydrolysis reactions which generate various monomeric species according to Eqs. (3)–(5); (c) aluminum hydrolysis products react with microalgae and form floculated particles (flocs); (d) the flocs become concentrated as microbubbles lift them to the surface (Eqs. (2) and (6)). In addition to electrolytic oxidation at the anode (Eq. (1)), aluminum ions can be generated at the cathode (Eq. (7) and (8)).

Anode:

\[ \text{Al} \rightarrow \text{Al}^{3+} + 3e^- \quad (1) \]
\[ 2\text{H}_2\text{O} \rightarrow \text{O}_2 + 4\text{H}^+ + 4e^- \quad (2) \]

In solution:

\[ \text{Al}^{3+} + 3\text{H}_2\text{O} \leftrightarrow \text{Al(OH)}_3^{2+} + \text{H}^+ \quad (3) \]
\[ \text{Al(OH)}_2^{3+} + 3\text{H}_2\text{O} \leftrightarrow \text{Al(OH)}_2^+ + \text{H}^+ \quad (4) \]
This electrolysis approach is particularly advantageous for marine species, as the electricity consumption was proved to be substantially reduced due to the high intense strength of seawater in comparison to freshwater species (Vandamme et al., 2011). In particular, Vandamme et al. (2011) demonstrated that marine microalgae required only one tenth of the power of freshwater species.

This study describes the design of continuous electrolytic microalgae (CEM) harvesting system that simplifies the cultivation and harvesting processes and allows for the continuous and efficient concentration of cells. The electrodes were subject to a polarity exchange (PE) in the middle of the operation to increase the system efficiency. PE improved the efficiency by eliminating the impermeable oxide film that often forms on electrodes, which prevents effective current transport between electrodes in conventional electrolytic processes (Vasudevan and Lakshmi, 2011). The PE process effectively creates two distinct phases: destabilization and flotation. Previous studies have shown that removal of microalgae was most efficient with a conventional electro-coagulation–flotation (ECF) process in two phases (Szpyrkowicz, 2005; Vandamme et al., 2011; Xu et al., 2010).

The general purpose of this work was to test the feasibility of a CEM harvest system with PE and to compare this new method with conventional electrolytic methods, ECF and electro-flotation (EF). The operating conditions for the CEM harvest system with PE were optimized by analyzing the effects of the applied current, initial pH, and mechanical mixing.

2. Methods

2.1. Microalgae strain, medium and chemicals

Nannochloris oculata (KMMCC-16), kindly provided by the Korean Marine Microalgae Culture Center (Busan, Korea), was used as the model marine microalgae. Modified f/2 medium (1 g yeast extract, 5 mg NaH2PO4H2O, 1 mL microelement solution, and 1 mL vitamin solution per 1 L of autoclaved seawater) was used for cultivation (Park, 2011). Table 1 lists the other primary cultivation conditions. The efficiency of a CEM harvest system depends on cell concentration, so microalgae cells were first harvested by centrifugation and then re-suspended with fresh seawater so that the same initial cell concentrations were used.

Table 1

<table>
<thead>
<tr>
<th>Conditions</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inoculation</td>
<td>0.1 g/L</td>
</tr>
<tr>
<td>Cultivation period</td>
<td>4 day</td>
</tr>
<tr>
<td>Light source</td>
<td>White LED</td>
</tr>
<tr>
<td>Light intensity</td>
<td>100 μmol/m²/s</td>
</tr>
<tr>
<td>Mixing rate</td>
<td>0.4 V/m</td>
</tr>
<tr>
<td>Temperature</td>
<td>25 °C</td>
</tr>
<tr>
<td>CO₂ concentration</td>
<td>2%</td>
</tr>
</tbody>
</table>

2.2. Electrolytic microalgae harvest chamber

All continuous electrolytic harvests were performed in a harvest chamber (inner length: 120 mm, inner width: 14 mm, inner height: 105 mm) which was connected to a cultivation tank. Fig. 1a shows a schematic diagram of the CEM harvest system. The harvest chamber was kept at a constant temperature by a water jacket. Initially, the cultivation tank and harvest chamber were filled with the same volume of re-suspended microalgae and fresh seawater. Then, the culture solution was circulated for 30 min before the onset of the continuous harvest, so that microalgae concentrations in the cultivation tank and harvest chamber were in equilibrium. Microalgae passed into and were harvested between the electrodes in that chamber. During the harvest, microalgae were floated in the harvest chamber and then cell-free seawater was circulated into the cultivation tank by peristaltic pumps (Masterflex L/S, Cole–Parmer USA) at a constant rate. Five minutes after termination of the CEM harvest, 10 mL of each sample was collected at three locations: the middle of the harvest chamber, the bottom of the harvest chamber, and at one point in the cultivation tank. Collection of samples at two locations in the harvest chamber allowed confirmation of cell recovery efficiency at two locations.

Two plate-type electrodes (width: 120 mm, height: 120 mm, thickness: 2 mm), an Al electrode (Mg: 4.0–4.9%, Mn: 0.4–1.0%, Cr: 0.05–0.25%), and DSE (Ti-RbO2) were placed 1.0 cm apart near the walls of the harvest chamber. Before each run, the Al electrode was immersed in a 5% HNO3 solution, mechanically ground with 600-grade abrasive paper, and then washed with acetone and distilled water so that its surface was uniform. The DSE electrode was immersed in 5% HNO3 solution and then rinsed with distilled water. All electrodes were dried prior to the experiments. The anode and cathode were connected to a DC power supply (S-3005Q, Fine Power Korea) in the galvanostatic mode. Consumed voltage was measured by a voltage meter (UT804, UNI-T China) during the entire electrolytic harvest period. A magnetic stirrer was used in the harvest chamber and cultivation tank. Table 2 shows the other primary harvest conditions. The CEM harvest system was operated in three modes (Fig. 1b): (1) Al coagulant addition from a sacrificial anode during the first half of harvest period (PE, polarity exchange), (2) Al coagulant addition from a sacrificial anode during the entire harvest period (ECF, electro-coagulation–flotation), and (3) no coagulant addition during the entire harvest period (EF, electro-flotation).

2.3. Subsequent cultivation of circulated culture after CEM harvest

After one set of CEM harvest experiments, the microalgae in the cultivation tank were centrifuged to separate circulated cells. Without further purification and sterilization steps, the circulated cells were re-suspended in circulated medium (CC + CM), or in fresh medium (CC + FM). Fresh cells and fresh medium (FC + FM) were used as a positive control. The three different batches had the same initial microalgae concentrations (0.1 g/L) and nutrient concentrations (modified f/2, described above).

2.4. Analytical methods

Chlorophyll concentration was measured as described by Boussiba and Vonshak (1991) and absorbance was measured at 666 nm by a UV–Vis spectrophotometer (Optizen 3220uv, Mecasys Korea). Based on chlorophyll concentration, the mean recovery efficiency (MRE) and recovery efficiency difference (RED) were calculated as:

\[
\text{MRE} = \left(1 - \frac{(A_{\text{m,harv}} + A_{\text{b,harv}})}{A_{\text{act}}} \right) \times 100\%
\]

(9)
\[
\text{RED} = \frac{\left| A_{m,\text{harv}} - A_{b,\text{harv}} \right|}{A_{\text{act}}} \times 100\% 
\]

where \( A_{m,\text{harv}} \) and \( A_{b,\text{harv}} \) are the chlorophyll concentrations in the middle \((m)\) and bottom \((b)\) of the harvest chamber of the CEM harvest system and \( A_{\text{act}} \) is the chlorophyll concentration of the actual processing microalgae during the operation of CEM harvest system (calculated by subtracting the final chlorophyll concentration from initial value in the cultivation tank). The electrical energy consumption (EEC), expressed as W h/g of recovered microalgae, was calculated as:

\[
\text{EEC} = \frac{U \times I \times t}{V \times C_{\text{act}} \times \text{MRE}/100} 
\]

where \( U \) is voltage (V), \( I \) is applied current (A), \( t \) is the time (h) of the harvest process, \( V \) is the volume (L) of the harvest chamber, \( C_{\text{act}} \) is the actual processing microalgal concentration (g/L), and MRE is the mean recovery efficiency (calculated from Eq. (9)). After the
CEM harvest system, the residual Al concentration was measured by ICP-OES (730-WA, Varian USA). The growth rate (GR) in the cultivation tank after the CEM harvest was calculated as described previously (Park, 2011):

\[
GR = \frac{(W_2 - W_1)}{(t_2 - t_1)} \text{ (g/L day)}
\]

where \(W_1\) and \(W_2\) are the dry cell weights (g/L) on day \(t_1\) and \(t_2\), respectively.

3. Results and discussion

3.1. Comparison of recovery efficiency of the polarity exchange (PE) and conventional electrolytic harvest (ECF and EF) modes in the CEM harvest system

The feasibility of using a PE mode in the context of the CEM harvest system was assessed by measuring the mean recovery efficiency (MRE, Eq. (9)), electrical energy consumption (EEC, Eq. (11)), and residual Al concentration (Table 3). The results indicated that the MRE of the PE mode was higher than that of the ECF mode. In conventional ECF, there are differences in the efficiency of chlorophyll removal at different vertical locations.

![Graphs showing mean recovery efficiency, residual Al concentration, and electrical energy consumption over time for different currents and harvest times.](image)

**Fig. 2.** Effect of current on mean recovery efficiency (a), residual Al concentration in the harvest chamber (b) and cultivation tank (c), and electrical energy consumption (d) at different harvest times. Conditions: pH = 8, temperature = 25 °C, stirring speed = 150 rpm.

<table>
<thead>
<tr>
<th>Harvest method</th>
<th>Electrical energy consumption (W h/g)*</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polarity exchange</td>
<td>1.08</td>
</tr>
<tr>
<td>Tangential flow filtration</td>
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</tr>
<tr>
<td>Polymer flocculation</td>
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<tr>
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<tr>
<td>Centrifugation</td>
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* Electrical energy consumption was calculated based on a MRE of 95.8% for polarity exchange.

Table 4

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* Electrical energy consumption was calculated based on a MRE of 95.8% for polarity exchange.
(Alfafara et al., 2002). Therefore, it was not possible for existing electrochemical technologies to completely separate cells from spent medium, a disadvantage for the subsequent steps of biodiesel production. Thus, the mean value of the recovery efficiency difference (REDs, Eq. (10)) of the PE and ECF modes were calculated over the time range 2.5–20 min to determine whether the PE mode enabled the uniform harvesting of cells at all levels. A low RED value indicates similar harvest efficiency in the middle and bottom and a large RED value indicates that there were more microalgae flocs at the bottom. A low RED (0.61%) was obtained upon application of the PE mode to the CEM harvest system. Use of ECF led to a RED of 11%, meaning that more cells were harvested in the middle than at the bottom. Alternatively, some microalgal flocs may have settled due to lack of buoyancy provided by ECF in the CEM harvest system. These initial results strongly suggest that the CEM harvest system provided higher and more uniform microalgae recovery in the PE mode than the conventional ECF mode. Fig. 1c shows floating microalgal flocs in the CEM harvest system with PE.

Although the PE mode allowed more effective of harvest of cells from almost all locations, the EEC of PE 15 was higher than that of ECF 15 and ECF 7.5 due to the use of the second phase of the PE mode. Immediately after the exchange of the anode and cathode, the voltage increased (data not shown). However, EEC of the CEM harvest system with PE was lower than other previously reported harvesting methods (Table 4).

PE 15 is a serial combination of ECF 7.5 and EF 7.5, but had more efficient Al utilization than the sum of ECF 7.5 and EF 7.5. About 62% and 20% of residual Al were reduced in each harvest chamber and cultivation tank by PE 15 in comparison to the sum of separate ECF 7.5 and EF 7.5. The use of increased Al resulted in 24% higher MRE for PE 15 than ECF 7.5. In the case of ECF 7.5, the microbubble density might not have been high enough for the formation and flotation of microalgal flocs. Moreover, PE 15 was more effective
than ECF 15, as indicated by the higher MRE and lower Al contamination. In the case of ECF 15, too much Al might have been introduced, resulting in a lower MRE and increased residual Al. The EF 15 process did not yield floating or settled microalgae flocs, possibly because the non-sacrificial anode did not generate Al. These results indicated that PE was well-suited for the CEM harvest system. In light of this success, the PE mode was used in all subsequent CEM harvest experiments.

3.2. Effect of current intensity on the CEM harvest system with PE

In all electrolytic harvest processes, the level of current is a critical determinant of Al coagulant dosage and microbubble density. DC currents of 0.25, 0.5, and 0.75 A were applied in the CEM harvest system to optimize the current in the PE mode. The results indicate that the time needed to reach 96% MRE declined as current increased (15 min at 0.25 A, 10 min at 0.5 A, 5 min at 0.75 A) (Fig. 2a). There were no increases in MRE beyond 96% for longer times at all three currents.

Residual Al concentration in the harvest chamber and cultivation tank increased with increasing current (Fig. 2b). In particular, the results indicate that residual Al concentration in the harvest chamber was 1.80 mg at 0.25 A, 3.16 mg at 0.5 A, and 2.52 mg at 0.75 A for an MRE of 96%. In addition, the rate of Al accumulation in the harvest chamber slowed down over time at all currents. However, the amount of accumulated Al in the cultivation tank was linearly proportional to harvest time (Fig. 2c). As the current increased, the rate of Al accumulation in the cultivation tank also increased due to the increased dissolution of Al. At an MRE of 96%, residual Al concentration in the cultivation tank was 2.50 mg at 0.25 A, 3.86 mg at 0.5 A, and 1.64 mg at 0.75 A.

The EEC at each current was found to be directly proportional to the length of the harvest time (Fig. 2d): 1.08 W h/g at 0.25 A, 1.68 W h/g at 0.5 A, and 1.48 W h/g at 0.75 A for a 96% MRE. Thus, considering the rate of Al accumulation and EEC at 96% MRE, the use of a low current (0.25 A) appeared to be more favorable in the CEM harvest system. Moreover, floating microalgae flocs had a green color at 0.25 A, although some flocs at higher currents (15 min at
0.5 A, 10 min at 0.75 A) were white (data not shown), possibly due to the formation of NaClO at high current (Gao et al., 2010a).

3.3. Effect of initial pH on the CEM harvest system with PE

The net charge of Al hydroxide is positive at acidic pH and negative at alkaline pH (Duan and Gregory, 2003). Thus, the formation of flocs, which is driven by the electrostatic interaction of microalgae and Al, depends on the pH of the solution. When sacrificial anodes are used in conventional electroly-coagulation processes, a low pH is favored because positively charged Al is attracted to negatively charged microalgae, leading to coagulation (Gao et al., 2010b; Vandamme et al., 2011).

The solution pH was found to affect the operation of the CEM harvest system operated with PE. Three pH values were tested (4, 6, and 8), revealing that pH 8 yielded the highest MRE within the first 5 min of operation (Fig. 3a). However, after 5 min, the MRE at all tested pH values converged on an MRE of ca. 93%. The RED was calculated at each pH in an attempt to explain these results. The amount of settled microalgae flocs declined as pH increased, as indicated by the RED at 2.5 min (3.83% at pH 4, 2.98% at pH 6, 2.14% at pH 8) and 5 min (2.66% at pH 4, 2.10% at pH 6, and 0.50% at pH 8). Flocs formed at lower pH may have been too heavy for the microbubbles to float. Thus, more algal flocs sank to the bottom of the harvest chamber and re-circulated to the cultivation tank, reducing cell recovery.

The residual Al concentration in the harvest chamber was lower at high pH (Fig. 3b) and the rate of Al accumulation in the cultivation tank was greater at lower pH (Fig. 3c). At the identical current, more Al was dissolved from the electrodes at a lower pH (Mouedhen et al., 2008), resulting in an increase in residual Al concentration.

Fig. 3d shows that EEC decreased as pH increased, indicating that higher pH would be more economical for a CEM harvest system. Considering the rate of Al accumulation and EEC at 96% MRE, a neutral pH appeared to be more favorable for the PE mode. Cultured microalgae could be used directly in the CEM harvest system without the need for additional pH control, which provided an additional advantage over conventional electrolytic processes. Maintenance of a pH of 7–9 during photoautotrophic cultivation of microalgae also helps to prevent cell lysis (Coutteau, 1996).

3.4. Effect of mechanical mixing on the CEM harvest system with PE

Some previous studies have examined the effect of mixing on microalgae harvest by electrolytic processes. Alfafara et al. (2002) showed that mechanical mixing resulted in better replacement of chlorophyll contents at the bottom with low current, probably due to the more efficient contact of algal flocs and microbubbles. On the other hand, Ilhan et al. (2008) reported that mechanical mixing interfered with the formation of microalgae flocs, and hence increased electricity consumption.

Mechanical mixing of the solution was found to affect the CEM harvest system operated with PE at two currents: 0.25 and 0.5 A. The results indicated that MRE was 7% greater at 0.25 A with mechanical mixing (Fig. 4a). At this low current, the number of microbubbles generated from the anode and cathode may have been insufficient for the flotation and formation of microalgal flocs. Thus, mechanical mixing might interact synergistically with microbubbles in causing microalgal flocs to float. In contrast, there was no noticeable effect of mechanical mixing on the MRE at 0.5 A (Fig. 4a). At 0.5 A, the number of microbubbles generated from the electrodes may have been sufficient for the formation and flotation of microalgae flocs. Accordingly, mechanical mixing may be unnecessary or even detrimental, because it may disrupt the algal flocs. Mechanical mixing at 0.5 A produced results that agreed well with the results of a previous study, which showed that the chlorophyll time course convergence was narrower at higher currents than at lower currents in the presence of mechanical mixing (Alfafara et al., 2002).

At 0.25 A with mechanical mixing, the residual Al concentration in the harvest chamber was 13% lower (Fig. 4b), possibly because mixing allowed more contact between Al and the microalgae. Without mechanical mixing at 0.5 A, the amount of settled Al was 31% and 8% lower in the harvest chamber and cultivation tank, respectively (Fig. 4c), possibly because of poor contact between Al and microalgae.

Mechanical mixing at 0.25 A or no mixing at 0.5 A yielded 11% or 2% lower EEC values, respectively (Fig. 4d). Thus, an appropriate adjustment of mixing speed and applied current may be able to improve the coagulation and flotation of algal flocs for more efficient microalgae harvesting in the CEM system with PE.

3.5. Cultivation of N. oculata in the cultivation tank after the CEM harvest system

One of the major advantages of the CEM harvest system is that it produces microalgal biomass in the cultivation tank simultaneously with electrolytic cell recovery in the harvest chamber. However, some toxic substances may result from cultivation (Roldolf et al., 2003) or harvesting (Chernou et al., 2011; Perreault et al., 2009) that would interfere with microalgae growth in the cultivation tank. The growth of circulated microalgae was assessed by measuring the microalgae population remaining in the cultivation tank after CEM harvesting.

Fresh medium with fresh cells (FM + FC), fresh medium with circulated cells (FM + CC), and circulated medium with circulated cells (CM + CC) were tested next. The appearances of cultures grown under these three conditions were similar in the first 12 h (Fig. 5). However, after 12 h, the CM + CC cultures had higher growth than the FM + FC and FM + CC cultures. At the end of the experiment, the CM + CC cultures had a growth rate (GR, Eq. (12)) of about 0.24 g/L day, and this was ca. 16% lower for FM + FC and FM + CC cultures. The increased algal growth in circulated medium might be due to the presence of certain stimulants (or absence of certain inhibitors) in this medium (Kim et al., 2011). Addi-
tionally, the appearance of the FM + FC and FM + CC cultures was unchanged during the whole growth period. These results indicate that N. oculata cells in the cultivation tank remain healthy during the harvest.

4. Conclusions

An innovative CEM harvest system for microalgae was developed by combining cultivation and harvesting steps. The addition of PE further improved this CEM harvest system, and operation in PE mode was more efficient than conventional electrolytic methods. In addition, the growth yield of circulated culture was higher than that of fresh culture, and recirculated cells were undamaged during CEM harvesting with PE. The present results suggest that PE applied in a CEM harvest system provides effective simultaneous microalgal cultivation and harvest, thereby reducing the cost of microalgal biodiesel production.

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References


