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Effective recovery of microalgal biomass using various types of emulsion polymers

Menghour Huy [a,1], Gopalakrishnan Kumar [a,*,1], Pooja Sharma [b], Ranjna Sirohi [c,d], Ashok Pandey [c,e], Sang-Hyoun Kim [f,**]

[a] Department of Chemistry, Bioscience, and Environmental Engineering, Faculty of Science and Technology, Forus, 8400 Stavanger, Norway
[b] Department of Environmental Microbiology, Babasaheb Bhimrao Ambedkar Central University, Vidya Vihar, Rae Bareli Road, Lucknow, Uttar Pradesh 226025, India
[c] Centre for Energy and Environmental Sustainability, Lucknow 226 001, Uttar Pradesh, India
[d] Department of Chemical and Biological Engineering, Korea University, Seoul, Republic of Korea
[e] Centre for Innovation and Translational Research, CSIR-Indian Institute of Toxicology Research, Lucknow 226 001, Uttar Pradesh, India
[f] School of Civil and Environmental Engineering, Yonsei University, Seoul 03722, Republic of Korea

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ABSTRACT

Microalgae biomass has been considered as one of the potential feedstocks in biofuel production. Yet, biomass harvesting poses a challenge to the overall production cost due to its low cell density. Flocculation has been marked as one of the promising processes in microalgae harvesting technology. In this study, the first screening of two anionic (A-230, and A-330E) and five cationic polymers (C-810E, C-810EL, C-810EB, C-810ELH, and C-810EMB) followed by gravity settling with the mixed microalgae concentration of 2.24 g_{TSS}/L revealed that anionic polymers are less effective. Whereas all cationic polymers achieved above 90% harvesting efficiency. Therefore, the maximum mass recovery of 98.7% with 86.8 g_{TSS}/L sediment content was achieved by adjusting pH to 6–0.6 mL/L (115.178 mg/$g_{biomass}$) of C-810E followed by 15-min settling. The cationic polymer addition followed by settling would enable cost-effective downstream processing of microalgal biomass.

1. Introduction

Energy demands awareness have grown worldwide as the development of clean and sustainable fuels to replace fossil fuels in the future perspective (EIA, 2017). Many types of biomass feedstocks have also been studied to produce clean biofuel. However, food vs fuels was still a big concern for biofuel generation while considering agricultural crops (Filip et al., 2017; Kuchler and Linnér, 2012). In the spotlight, algae have the capability of giving high biomass productivities with high lipid content by CO_2 consumption leading to an environmental-friendly approach have earned more attention from many researchers (Ananthi et al., 2021b; Dassey and Theegala, 2013; Farooq et al., 2022; Huy et al., 2018). Somehow, it has been marked as not economically regarded due to its overall process cost. In order to obtain final cost-effective products, harvesting plays a major role in economic consideration for the whole operation.

Many harvesting technologies have been invented and carried out such as coagulation, flocculation, and centrifugation. Therefore, selecting cost-effective technology is still challenging (Ananthi et al., 2021a; Behera et al., 2015; Japar et al., 2017; Laamanen et al., 2016). Some harvesting technologies can only be done within a lab-scale and could not apply to the full-scale operation because of the operational cost (Mubarak et al., 2019). Overall harvesting technologies reported centrifugation is a method capable of acquiring high-value biomass without contamination but it's countered by the cost of the operation (Dassey and Theegala, 2013; Uduman et al., 2010; Wang et al., 2019). Some other researchers introduce flotation which efficiently harvests microalgae biomass by cell flotation activities (Laamanen et al., 2016). Cell flotation and dispersed ozone flotation promise to minimize the cost of operation with floatation technology (Ndikubwimana et al., 2016). However, cell floatation needs to be operated along with other processes to complete biomass harvesting (Ghazvini et al., 2022). For instance, the combination between coagulation and flotation together would bring almost 100% of biomass recovery. However, the cost of the overall

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* Correspondence to: University of Stavanger, Norway.

** Corresponding author.

E-mail addresses: Gopalakrishnan.kumar@uis.no (G. Kumar), sanghkim@yonsei.ac.kr (S.-H. Kim).

[1] Authors have equal contribution to this work.

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Table 1
Chemical/Physical Characteristic of the polymers.

Name	Polymer content (%, w)	Ionicity	Bulk viscosity (cps)	Specific gravity at 25°C	Chemical name
A-230E	29	Medium	250 – 450	1.02 – 1.06	Sodium 2-Propeoate 2-Propenamide Polymer
A-330E	29	Medium	350 – 950	1.02 – 1.06	
C-810E	43	High	250 – 450	1.02 – 1.06	Ethanaminium,N,N,N-trimethyl-2-((1-oxo-2-propenyl)oxy)-chlorid), polymer with 2-propenamide
C-810EL	43	High	100 – 300	1.02 – 1.06	
C-810EB	46	High	300 – 500	1.02 – 1.06	
C-810ELH	43	High	50 – 250	1.02 – 1.06	
C-810EMB	46	High	300 – 500	1.02 – 1.06	

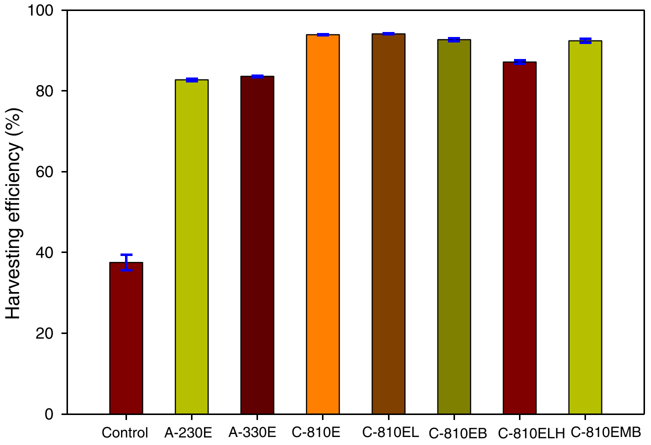

Fig. 1. Harvesting efficiency of each polymer at 0.5 mL/L dosage and 1-hr settling.

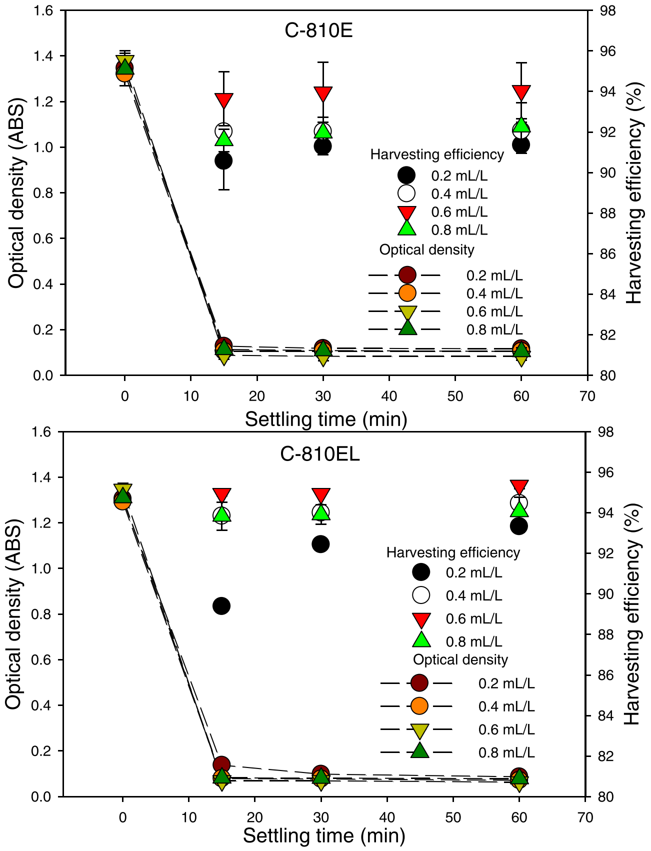

Fig. 2. Harvesting efficiency according to settling time at various dosages of C-810E and C-810EL.

operation is still not favorable (Xia et al., 2017).

Current studies on harvesting using flocculation followed by sedimentation could significantly bring low cost with the simple operation (Wan et al., 2015). Various flocculation technologies have been developed and applied such as inorganic flocculation, organic flocculation, and bio-flocculation (Ananthi et al., 2021a; Mubarak et al., 2019; Ummalyma et al., 2017; Wan et al., 2015). Moreover, microalgae surface charge was considered as negatively surrounding its cell (Gultom and Hu, 2013; Shelef and Sukenik, 1984). For efficient harvesting, a positively charge supplier would be needed to influence the flocculation activity of microalgae. Over the literature study, bio-flocculants have

Table 2
Comparison of flocculant dosage and harvesting efficiency with various flocculants.

Species	Flocculants/coagulants	Dosage	Microalgae biomass	Dose per gram biomass	Harvesting efficiency	Reference
		mg/L	g/L	mg/g $_{\text{biomass}}$	%	
Mixed microalgae	C-810E	258	2.24	115.178	98.7 ± 0.6	This study
Mixed microalgae	C-810EL	258	2.24	115.178	94.1 ± 0.1	This study
Chlorella vulgaris	PAETAC	–	0.31	102	~86	Nguyen et al. (2022)
Chlorella vulgaris	PAmPTAC	–	0.31	252	~86	Nguyen et al. (2022)
Chlorella vulgaris	Chitosan	250	1.2	208.33*	91.9	Zhu et al. (2018)
Chlorella vulgaris	$Al_2(SO_4)_3$	2500	1.2	2083.33*	92.4	Zhu et al. (2018)
Chlorella vulgaris	$Al_2(SO_4)_3$	180	0.36	504	77	Vu et al. (2020)
Chlorella vulgaris	$FeCl_3$	160	0.36	448	86	Vu et al. (2020)
Chlorella vulgaris	Chitosan	200	0.36	560	62	Vu et al. (2020)
Chlorella vulgaris	$FeCl_3$ + Chitosan	$FeCl_3$: 40 Chitosan: 80	0.36	$FeCl_3$: 111 Chitosan: 224	81	Vu et al. (2020)
Chlorella vulgaris	$Al_2(SO_4)_3$ + Chitosan	$Al_2(SO_4)_3$: 40 Chitosan: 80	0.36	$Al_2(SO_4)_3$: 111 Chitosan: 224	89	Vu et al. (2020)
Scenedesmus sp.	$Al_2(SO_4)_3$	500	1.2	416.67*	95	Oliveira et al. (2018)
Scenedesmus sp.	Polyacrylamide (PAM)	75.5	0.54	139.81*	90	Wu et al. (2015)

[*] Calculated by Dosage/Microalgae biomass.

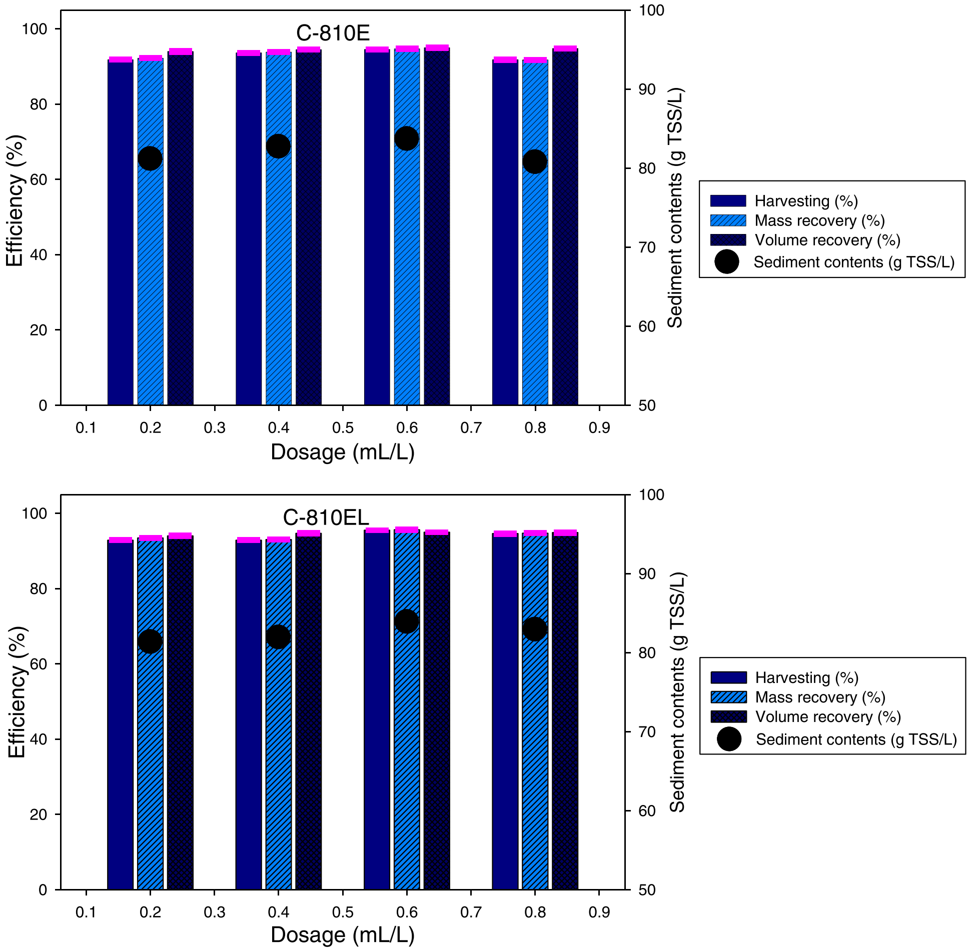

Fig. 3. Harvesting, mass recovery, and volume recovery efficiencies and sediment TSS content at various dosages of C-810E and C-810EL with 15-min settling.

been studied to precipitate microalgae biomass by using another kind of biomass as flocculants, but the requirement of a high dosage for the biomass flocculants would limit the application in full-scale (Ummalyma et al., 2017; Vu et al., 2020; Zhu et al., 2018).

This study investigated effective harvesting with polymer addition and gravity settling at various initial pH values and dosages. Although the mass recovery and the solids content of sediment are essential to determine the cost of microalgal-based product development, most of the previous research reported the reduction of optical density value in the supernatant, harvesting efficiency, only. In this study, the mass recovery and the solids content of sediment were also evaluated towards accuracy and efficiency.

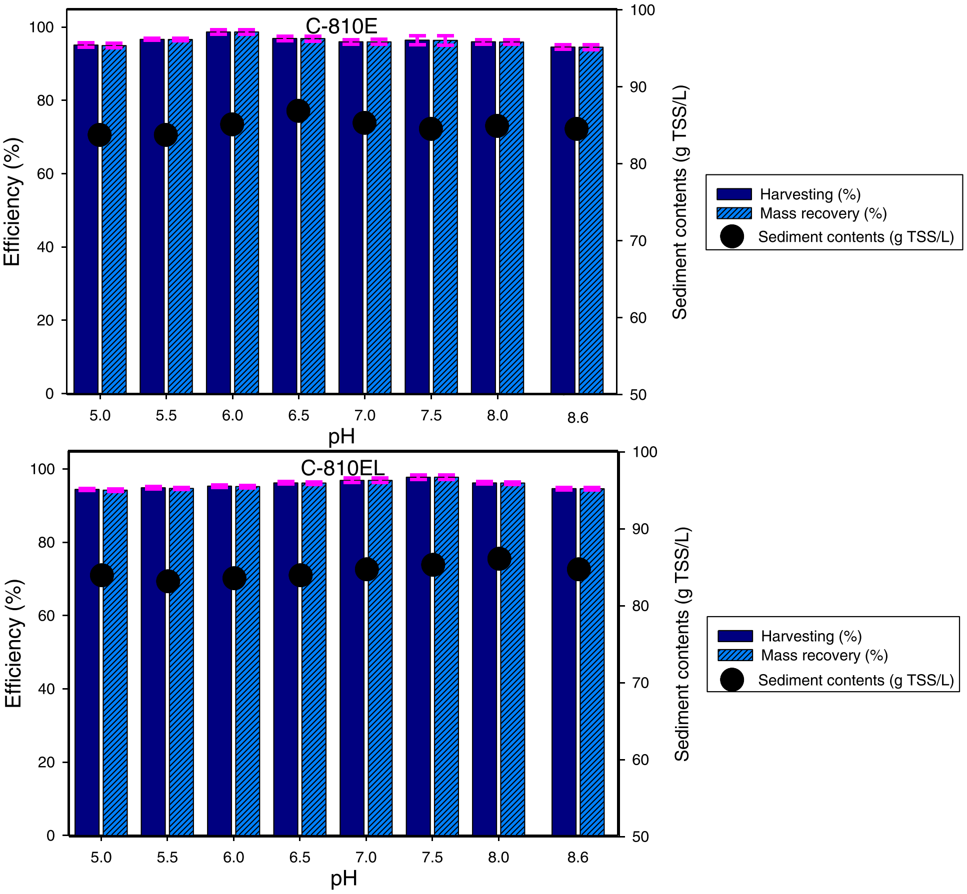

Fig. 4. Harvesting and mass recovery efficiencies and sediment TSS content at 0.6 mL/L of C-810E and C-810EL with 15-min settling under various pH values.

2. Materials and methods

2.1. Collection and cultivation of mixed microalgae consortia

Microalgae consortia were isolated from an opened wastewater pond in Daegu University, South Korea. The culture was collected inside a 5-L sampling bag and then stored in a 4 °C cold temperature room. The collected sample was observed under microscopy to identify the microalgae species present in it. Furthermore, five sequencing subculturing of microalgae cultivations were carried out with Bold Basal's medium which was commonly used for freshwater microalgae. The mixed consortia was highly dominated by *Chlorella sp.* and followed by *Scenedesmus sp.*

In order to obtain enough biomass for the further experiment, mixed microalgae culture was cultivated in a lab-scale photo-bioreactor. The lab-scale cultivation was designed to consist of a 50-L working volume equipped with 170 μmol.m^{-2}.s^{-1} light intensity provided by two LED lights (Samsung LED Light 30 W, model FPL-55 W white color, Korea). It was performed in 12 h/12 h of dark/light and ambient air was provided by an aeration pump (AMAZONPET, model SH-A2, China) for microalgae photosynthesis. The cultivation was carried out for 34 days at room temperature until the mixed microalgae's growth was reached 2.24 g_{TSS}/L at the stationary phase.

2.2. Experimental setup

In this study, mixed microalgae consortia were harvested by the various types of polymers in emulsion form. Two kinds of sodium 2-propepeoate 2-propenamide polymer (A-230E and A-300E) were used for anionic flocculants, while five kinds of ethanaminium,N,N,N-trimethyl-2-((1-oxo-2-propenyl)oxy)-chlorid), polymer with 2-propenamide (C-810E, C-810EL, C-810EB, C-810ELH, and C-810EMB) were used as cationic flocculants. The emulsion polymers were obtained from Eyang Chemical Co., Ltd, South Korea. The viscosity ranges from 500 to 2000 cps. The ionicities of the polymers were middle anionic for anionic polymers and high cationic for cationic polymers. Overall characteristics could be seen in Table 1.

The experiments were categorized into 3 stages: (i) screening the efficient polymers among various types of polymers, (ii) dosage optimization for the chosen polymer, and (iii) enhancement of the polymer flocculants by pH adjustment. All the flocculation experiments were conducted at room temperature. Flocculation was conducted by adding the polymer to the microalgae biomass at 120 rpm for 2 min (Supplementary Fig. 1, Fig. S1). The first experiment was performed with a fixed dosage of 0.5 mL/L with all polymers. After selecting the effective polymer, different dosages of 0.2, 0.4, 0.6, and 0.8 mL/L were investigated. Polymer concentration correlation with these dosages can be seen in Supplementary Table S1. With the efficient polymers and optimal dosage, the effect of the pH was examined by adding 2 N of HCl and 1 N of KOH to reach the pH values of 5.0, 5.5, 6.0, 6.5, 7.0, 7.5, 8.0, and 8.6.

2.3. Analytical procedures and calculation

Total solids (TS), volatile solids (VS), total suspended solids (TSS), volatile suspended solids (VSS), and chemical oxygen demands (COD) were analyzed followed Standard Methods (APHA/AWWA/WEF, 1998). Optical density (OD) was measured by UV-Spectrophotometer (SHIMADZU, UV–VIS mini 1240, Japan) at 750 nm.

2.4. Data analysis

All experiments were carried out in triplicates. The statistical ana-

Table 3
Comparison of pH and harvesting efficiency of microalgal biomass using flocculants/coagulants.

Species	Flocculants/ coagulants	pH	Harvesting efficiency (%)	Reference
Mixed microalgae	A-230E	8.58	82.7 ± 0.3	This study
Mixed microalgae	A-330E	8.58	83.6 ± 0.2	This study
Mixed microalgae	C-810E	8.58	93.9 ± 0.2	This study
Mixed microalgae	C-810E	6.0	98.7 ± 0.6	This study
Mixed microalgae	C-810EL	8.58	94.1 ± 0.1	This study
Mixed microalgae	C-810EL	7.5	97.8 ± 0.6	This study
Mixed microalgae	C-810EB	8.58	92.7 ± 0.4	This study
Mixed microalgae	C-810ELH	8.58	87.1 ± 0.5	This study
Mixed microalgae	C-810EMB	8.58	92.4 ± 0.5	This study
Parachlorella	Cationic starch	N.D	> 90	Vandamme et al. (2010)
Chlorella protothecoides	Cationic starch	7.7, 10	98	Letelier-Gordo et al. (2014)
Scenedesmus sp.	Polyamine polymer	N.D.	> 90	Gupta et al. (2014)
Scenedesmus sp.	Alum	7	92.3	Gupta et al. (2014)
Scenedesmus sp.	Chitosan	7	> 90	Gupta et al. (2014)
Chlorella vulgaris	PDADMAC	N.D.	> 90	Gerchman et al. (2017)
Scenedesmus sp.	$FeCl_3$	7	98.8	Das et al. (2016)
Chlorella sp.	Morina oleifera seed	N.D.	95	Abdul Hamid et al. (2014)

lyses were conducted using Microsoft Office (Excel 2013) and shown as mean value ± standard deviation.

The performance of solid-liquid separation was evaluated using two separation parameters, harvesting efficiency (Das et al., 2016) and mass recovery efficiency (Hjorth et al., 2009) as below.

$$Harvesting\quad Efficiency\quad (\%) = \left(1 - \frac{OD_f}{OD_i}\right) \times 100 \tag{1}$$

while,OD_i = OD at 750 nm before separation.

OD_f = OD at 750 nm in the supernatant

$$Mass\quad recovery\quad effiency\quad (\%) = \frac{m_{\mathrm{TSS},sediment}}{m_{TSS,slurry}} \times 100 \tag{2}$$

While, $m_{\mathrm{TSS,\ slurry}}$ = mass of TSS before separation (g) and $m_{\mathrm{TSS,sediment}}$ = mass of TSS in sediment (g).

3. Results and discussion

3.1. Harvesting efficiency according to polymer and its dosage

Fig. 1 compares the harvesting efficiency of each polymer. Microalgae settled by gravity force only (control) showed 37.5 ± 1.9% of the harvesting efficiency after 1 hr settling, which ensured that the use of an enhanced method is inevitable for harvesting microalgae by gravity settling. In the same figure, the peak harvesting efficiencies of 93.9 ± 0.2 and 94.1 ± 0.1% were obtained by two kinds of cationic polymers for the C-810E and C-810EL, respectively, with a dosage of 0.5 mL/L. Other cationic polymers, C-810EB, C-810ELH, and C-810EMB, achieved harvesting efficiencies of 92.7 ± 0.4, 87.1 ± 0.5, and 92.4 ± 0.5%, respectively. On the other hand, the anionic polymers, A-230E and A-330E showed harvesting efficiencies of 82.7 ± 0.3 and 83.6 ± 0.2%, respectively, which revealed that anionic flocculants are less effective than cationic flocculants on harvesting purposes of microalgae.

Fig. 2 indicated that 15 min was enough for settling with C-810E and C-810EL. It also illustrates that each dose resulted in above 89% of harvesting efficiency where the optimum dosage for both cationic polymers was 0.6 mL/L. A higher dosage than the optimum could not enhance but slightly decrease the harvesting efficiency. This may happen due to the excess polymer suspended in the supernatant causing this drop in harvesting efficiency. This excess might result in the positive zeta potential which could hinder flocculation and settling (Ummalyma et al., 2017). Therefore, this overdose excess of cationic flocculant was also reported by the other study using cationic starch (Greenfloc 120) to recover *Parachlorella* biomass was given a similar pattern where the optimal efficiency increased to more than 90% at a certain range of flocculant concentration, where higher dose than reported optimal declines the efficiency (Vandamme et al., 2010). The counterproductive effect of the excess overdose of several other polymer flocculants was also reported by other studies (Gerchman et al., 2017; Gupta et al., 2014; Nguyen et al., 2022, 2019). This phenomenon of an overdose of flocculant was found and reported as dispersion destabilization (Liu et al., 2009; Vandamme et al., 2010).

Over the literature survey on the dosage of flocculant per gram microalgae biomass, Nguyen et al. (2022) reported two cationic polymers were used to harvest *Chlorella vulgaris* resulting in about 86% efficiency required 102 mg/g and 252 mg/g for poly[2 (acryloyloxy) ethyl]trimethylammonium chloride (PAETAC) and poly(3 acrylamidopropyl)trimethylammonium chloride (PAmPTAC), respectively. In one other study, Zhu et al. (2018) reported that in order to harvest above 90% of *Chlorella vulgaris* biomass using chitosan and aluminum sulfate required 0.25 and 2.5 g/L, respectively. The results were reported with harvesting biomass concertation of 1.2 g/L which corresponds to 208.33 $\mathrm{mg/g_{biomass}}$, and 2083.3 $\mathrm{mg/g_{biomass}}$ for chitosan and aluminum sulfate, respectively. Generally, using inorganic salts as flocculants requires high dose with the drawback of contamination of harvested microalgae biomass (Mubarak et al., 2019; Oliveira et al., 2018; Zhu et al., 2018). Interestingly, Vu et al. (2020) investigated the synergistic effect between inorganic salts (aluminum sulfate and ferric chloride) with chitosan for harvesting *Chlorella vulgaris*. As a result of charge neutralization and bridging mechanism between the dual flocculants, the study suggested the combination of aluminum sulfate with chitosan will lower the cost by 30% compared to the single-use of chitosan. The application of chitosan as a flocculant is regarded as a promising candidate for biofuel production due to its biodegradability but its cost challenges scalability (Mubarak et al., 2019; Vu et al., 2020; Yin et al., 2021). Table 2 shows the summary of the reported dosage of several flocculants for microalgae harvesting. Since the result of this study shows lower flocculants dose per gram biomass, these selected flocculants can be foreseen as potential candidates for harvesting microalgae biomass. However, further study of the effects of these polymers in the downstream process is required.

3.2. Mass recovery efficiency and sediment solids content

Apart from the harvesting efficiency, mass recovery efficiency would also state the distribution of solids compounds toward the sediment and supernatant fractions inside the sample where the greater the mass recovery efficiency expressing the higher amount of the compound presented (Hjorth et al., 2009). Also, the solids content of the sediment is another important parameter to determine the transport, storage, and processing costs of downstream units. Therefore, the volume recovery efficiency provided extra information on water recovery efficiency from this harvesting process. Fig. 3 depicts the microalgal biomass recovery efficiency analyzed from TSS content and volume of supernatant and sediment. The peak mass recovery efficiencies of 94.7 ± 0.4 and 95.8 ± 0.4% were found at 0.6 mL/L addition of C-810E and C-810EL, respectively. At the optimum condition without pH adjustment, TSS

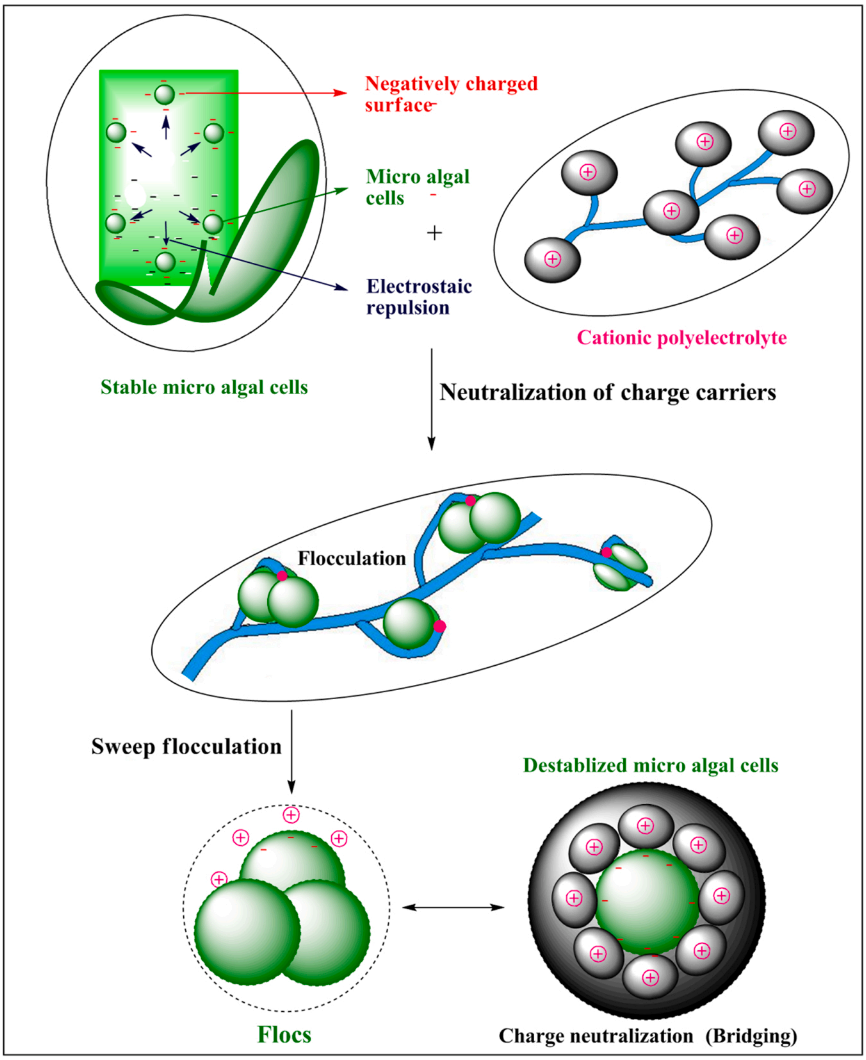

Fig. 5. Mechanism of flocculation of microalgal cells via polymers.

contents in the sediment were 83.7 ± 0.4 and 83.9 ± 0.2 g_{TSS}/L for C-810E and C-810EL, respectively. It implied that the cationic polymer addition followed by 15-min settling generated about 40 times more concentrated microalgal biomass flow than un-harvested culture. At 0.8 mL/L dosage, the mass recovery efficiency dropped to 91.8 ± 0.4 and 94.7 ± 0.3% for C-810E and C-810EL, respectively. With the same negative effects of excess polymers as harvesting efficiency, this confirmed that 0.6 mL/L was the optimum dosage for both polymers which lead to the optimization study by pH adjustment. In the life cycle assessment (LCA) study of Yuan et al. (2015), dewatering biomass is one of the factors affecting the cost of operation to concentrate solids level of biomass before going further downstream processing. The study suggested Biofloc with dissolved air flotation model for harvesting and dewatering microalgae biomass, which was able to concentrate solid to 50 g/L from bulk biomass for harvesting, then dewatering process (centrifugation) concentrated it to 180 g/L for the need for further process (Yuan et al., 2015). The suggested harvesting and dewatering process is significantly cost-effective compared to a single centrifugation process. Thereby, since the result of our study demonstrated higher concentrated biomass in the harvesting process, this will then improve the energy consumption for the dewatering process by the reduction of biomass loading rate.

3.3. *Optimization of mass recovery efficiency by pH adjustment*

In the flocculation and coagulation process, pH is one of the main parameters to determine separation efficiencies. Therefore, the effect of initial pH was investigated from pH 8.6 (original value) to 5.0. Fig. 4 illustrated the variation of harvesting and mass recovery efficiencies through pH adjustment. For C-810E, the mass recovery efficiency was improved by decreasing pH to 6, where 98.7 ± 0.6% of biomass was recovered. The sediment TSS content was also enhanced to 86.8 ± 0.4 g/L. However, the mass recovery efficiency started to drop when pH was at 5.5 and 5, as 96.6 ± 0.4 and 95.0 ± 0.6%, respectively. The result pointed out that pH 6 was favorable for harvesting microalgae biomass with cationic polymer C-810E. The mass recovery efficiency of C-810EL reached its highest at 97.7 ± 0.6% with 86.1 ± 0.4 g TSS/L at pH 7.5. Overview on a literature survey, Gupta et al. (2014) reported that at similar pH, harvesting *Scenedesmus sp.* with alum gave 92.3% at pH 7 and over 90% of biomass recovery at all pH with a low dosage of polyamine polymer (Gupta et al., 2014). In one other study, Das et al. (2016) reported that harvested *Scenedesmus sp.* by using $FeCl_3$ and adjusted pH to 7.0 gave 98.8% of harvesting efficiency, and lower pH to 3 by H_2SO_4 was quite efficient as 99.5% was achieved (Das et al., 2016). At the same pattern with mass recovery efficiency, harvesting efficiency was as well confirmed as 98.7 ± 0.6 and 97.7 ± 0.7% for C-810E (pH 6) and C-810EL (pH 7.5) were recorded, respectively. A similar trend of this decreasing pH gives the enhancement of flocculation efficiency was also examined and reported by Liu et al. (2013). This self-flocculation of microalgae by pH adjustment for biomass harvesting is one of the promising low-cost technologies but it hardly meets the demand for commercial criteria (Wan et al., 2015). Liu et al. (2013) reported in their study that the significantly improved flocculation activities of microalgae are at pH adjusted to 4.0 or below, which will provide more than 90% of flocculation efficiency. Therefore, it was also observed that microalgae begin to flocculate with pH decrease from pH 6.7 but the flocs still remain suspended in the growth medium. By the phenomenon of self-flocculation by this pH adjustment may explain the slight increase of mass recovery by the concentration of added HCl (C-810E adjusted to pH 6) inducing the flocculation activity of microalgae cell and suspended in the growth medium. The resulted larger floc then binds with the added cationic polymers to help settle more biomass. The other similar optimal pH for cationic polymers to C-810EL (pH 7.5) was also reported by the other study, where 7.7 was a promising pH to recover *Chlorella protothecoides* by using the Greenfloc 12 (Letelier-Gordo et al., 2014). Table 3 compares the harvesting efficiency for microalgae using various polymers and pH values.

The mechanism of microalgal cells flocculation has been shown in Fig. 5. As represented the chemical reactions involved are categorized as follows,

a. Neutralization of charge carriers
b. Sweep flocculation and
c. Gravity filtration

The algal cell particles are typically negatively charged in solution and form stable colloids so the cationic acrylamide-based polymers work very effectively via their inductive and electromeric effects. The existence of amide functional (–C=O–NH$_2$) and other electronegative atoms in the matrix enhance a negative surface charge to microalgal cells because the lone pair of the electron from –N atom of the amide shifted towards more electro negative –O atom. Nevertheless, the cationic polymers C-810E and C-810EL showed favorable mass recovery performance even without pH adjustment, which would be applicable, given that the following downstream process requires mass recovery and sediment solids content up to 95% and 80 g/L, respectively. Therefore, the cost of pH adjustment should be considered in the industrial-scale application.

4. Conclusions

Harvesting microalgae with cationic polymer could be one of the viable options for the industrialization of microalgal biomass cultivation due to high mass recovery efficiency and solids content in the sediment. These results demonstrated the potential of energy consumption reduction in harvesting and dewatering which promised to reflect the overall production cost. The maximum mass recovery of 98.7% was achieved with the sediment of 86.8 g TSS/L by the addition of C-810E at 0.6 mL/L at pH 6.0 followed by 15-min settling, while with the same dose, C-810EL achieved 95.5% mass recovery with 86.1 g TSS/L without pH adjustment and favorable efficiency was attained at initial pH of 8.6. This indicated that the cost related to pH adjustment with this slight enhancement should be strongly taken into account in the industrial scale consideration. Therefore, further research on the effect of these polymers on the later production chain is needed.

CRediT authorship contribution statement

Menghour Huy: Conceptualization; Data curation, Writing – original draft; Writing – review & editing. **Gopalakrishnan Kumar:** Conceptualization, Data curation, Formal analysis, Funding acquisition. **Pooja Sharma:** Validation, Visualization, Writing – review & editing. **Ranjna Sirohi:** Validation, Visualization, Writing – review & editing. **Ashok Pandey:** Writing – original draft; Writing – review & editing. **Sang-Hyoun Kim:** Conceptualization, 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.

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Appendix A. Supporting information

Supplementary data associated with this article can be found in the online version at doi:10.1016/j.jbiotec.2022.08.010.

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