



A comparison of zeta potentials and coagulation behaviors of cyanobacteria and algae

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ABSTRACT

In this study, the zeta potential of *Microcystis aeruginosa* (cyanobacteria), *Synechococcus* sp. (picocyanobacteria) and *Chlorella vulgaris* (algae) was investigated in order to determine the zeta potential range for optimum cell removal. Algae and cyanobacteria species were treated by coagulation–sedimentation using 0–500 mg/l polyaluminum chloride (PACl). Analyses included zeta potential measurement, cell counts and turbidity removal. The role of pH on the zeta potential also investigated in this study. The pH of coagulation system was adjusted to pH 6.5 and 7.0. At pH 6.5, the optimum zeta potential bands were between –2.1 mV and +20.45 mV for *Microcystis aeruginosa*, +3.45 mV and +8.71 mV for *Chlorella vulgaris* and +7.41 mV and +13.33 mV for *Synechococcus* sp. The ranges were much narrower at pH 7 than at pH 6.5. The cell removal efficiencies were 98.9%, 90.6% and 55.7% for *Microcystis aeruginosa*, *Chlorella vulgaris* and *Synechococcus* sp, respectively. The implications of such findings are that the charge measurement can be used for controlling coagulation of algae and cyanobacteria. In addition, the type of cell species in the system was shown to a significant factor in the coagulation performance.

Keywords: Zeta potential; Coagulation; Turbidity; Cyanobacteria; Algae

1. Introduction

Algae are ubiquitous in rivers, lakes, dams and reservoirs which supply water treatment plants. With the dramatic seasonal increase in the algae population, there is a corresponding increase in cyanobacteria, which can be attributed to the increase in algal blooms. The increase in cyanobacteria and algae population causes some problems in the drinking water treatment process, including high turbidity, high coagulant demand, fragile and poor flock formation. The water treatment process efficiency can be impaired as a result [1].

The coagulation–flocculation process is commonly employed in water treatment facilities for algae and

cyanobacteria removal, but this process is inadequate when the algae and cyanobacteria population increase. If the coagulation process is unsuccessful, algae–cyanobacteria cells and coagulant left over from the coagulation process can penetrate to downstream filters often resulting in filter clogging or breakthrough. Residual algae cells and their compounds can form disinfection by-products (DBP), such as trihalomethanes (THM), or can contribute to giving the water an undesirable taste and odor, or result in the formation of toxic metabolites [2–4]. Moreover, it has been shown that cyanobacteria, especially pico-sized (0.2–2.0 μm) cyanobacteria such as *Synechococcus* sp, contribute to the formation of assimilable organic carbon (AOC) which promotes bacterial regrowth in distribution systems [5,6]. For all these reasons, the removal

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of cyanobacteria and algae without cell damage prior to both the filtration and disinfection process is desirable to improve the quality of treated water. However, the removal of algae and cyanobacteria is more difficult than removing suspended solids because their physical and chemical characteristics vary widely; not only do they vary in size, but their density and surface charge also differ.

The successful removal of algae and cyanobacteria cells by coagulation and flocculation significantly depends on the stability of the system, which can be affected by surface charge. The surfaces of algae and cyanobacteria cells are negatively charged either as a result of dissociation, the ionization of surface functional groups or adsorption of ions originating from organic matter. Measurement of surface charge of cyanobacteria and algae gives zeta potential. A reduction in the magnitude of the negative zeta potential signifies a reduction in the repulsive electrostatic forces. When the attractive van der Waals forces overcome these electrostatic forces, the critical zeta potential is reached and then organic and inorganic particles agglomerate. This agglomeration of particles constitutes the basis of the coagulation process [7,8]. As such, the zeta potential is an important parameter when investigating the coagulation mechanism.

In last 20 years, the interaction between the zeta potential and the coagulation of organic and inorganic materials, including natural organic matter (NOM) and colloids, has been heavily investigated. The optimum zeta potential range has been reported to be between -10 mV and $+5$ mV, and -20 mV and $+5$ mV for NOM and kaolin, respectively [9]. Sharp et al. [10] reported that a threshold range between -10 mV and $+3$ mV represents a safe guide to appropriate zeta potential for optimized NOM treatment. Few studies, however, have been done on the use of the zeta potential for estimating the optimum coagulant dose and monitoring the coagulation of algae and cyanobacteria. While it was claimed that the zeta potential range between -14.5 mV and $+12$ mV for optimum removal efficiency of algae (*Chlorella vulgaris*), since this conclusion was based on the results of studies investigating the coagulation–flocculation–flotation process, the results may not be applicable to the coagulation–flocculation–sedimentation process [2]. Therefore, studies need to be done to clarify the operational zeta potential range of various algae and cyanobacteria species for better control of the coagulation process. To date, numerous studies have been conducted on the removal of cyanobacteria larger than 3.5 μm by the coagulation process; however, no studies have been done on the removal of pico-cyanobacteria such as *Synechococcus* sp. despite clear indications that pico-cyanobacteria contribute to the turbidity of treated water [11].

Hence, the purpose of the current study was to investigate the applicability of the zeta potential to control the coagulation of different algal systems. In this study, we compared *Microcystis aeruginosa*, *Chlorella vulgaris* and *Synechococcus* sp. These species are commonly found in water sources and are believed to be associated with water quality and treatment problems in water treatment plants.

2. Materials and methods

2.1. Cyanobacteria and algae cultivation

The freshwater cyanobacteria cultures *Microcystis aeruginosa* (NIES-87) and *Synechococcus* sp. (NIES-1348) and algae *Chlorella vulgaris* (NIES-2170) were provided by the National Institute for Environmental Studies (NIES) in Japan. *Chlorella vulgaris* was cultivated in an axenic C medium. The C medium was prepared according to the standard of the NIES medium preparation and was composed of 15 mg $\text{Ca}(\text{NO}_3)_2 \cdot 4\text{H}_2\text{O}$ (CAS 13477-34-4), 10 mg KNO_3 (CAS 7757-79-1), 5 mg $\beta\text{-Na}_2$ glycerophosphate- $5\text{H}_2\text{O}$ (CAS 819-83-0), 4 mg $\text{MgSO}_4 \cdot 7\text{H}_2\text{O}$ (CAS 10034-99-8), 0.01 μg Vitamin B_{12} (CAS 68-19-9), 0.01 μg Biotin (CAS 58-85-5), 1 μg Thiamine HCl (CAS 67-03-8), 50 mg Tris (hydroxymethyl)aminomethane (CAS 83147-39-1), 0.3 mg/L $\text{Na}_2\text{EDTA} \cdot 2\text{H}_2\text{O}$ (CAS 6381-92-6), 0.0588 mg/L $\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$ (CAS 10025-77-1), 0.0108 mg/L $\text{MnCl}_2 \cdot 4\text{H}_2\text{O}$ (CAS 13446-34-9), 0.0031 mg/L $\text{ZnSO}_4 \cdot 7\text{H}_2\text{O}$ (CAS 7446-20-0), 0.0012 mg/L $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$ (CAS 7791-13-1), 0.00075 mg/L $\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$ (CAS 7631-95-0). *Microcystis aeruginosa* and *Synechococcus* sp. were cultivated in an axenic CB medium. While the CB medium was being prepared, Bicine (CAS 17123-43-2) was added instead of Tris (hydroxymethyl) to the C medium. The pH of both mediums was adjusted to pH 9.0 by adding 0.1 M NaOH (CAS 95077-05-7) or 0.1 M HCl (CAS 9004-54-0). *Microcystis aeruginosa*, *Synechococcus* sp. and *Chlorella vulgaris* were grown in a 200-ml Erlenmeyer flask at a unialgal level with 100 ml of medium on a rotary shaking device 90 rpm with constant illumination ($65 \mu\text{mol}/\text{m}^2/\text{s}$) and a 12 h/12 h light/dark cycle in an incubator. The temperature was maintained at 25°C . Cell populations were measured by counting at least 100 cells in triplicate using a light microscope. According to suggestions in previous studies that the stage of life cycle can influence the zeta potential [12] the cells during the onset of the stationary phase of culture were used in the coagulation experiments since the cell population density was at its highest at this stage (Fig. 1). The stationary phase concentration, cell size and the surface area of *Microcystis aeruginosa*, *Synechococcus* sp. and *Chlorella vulgaris* are shown in Table 1.

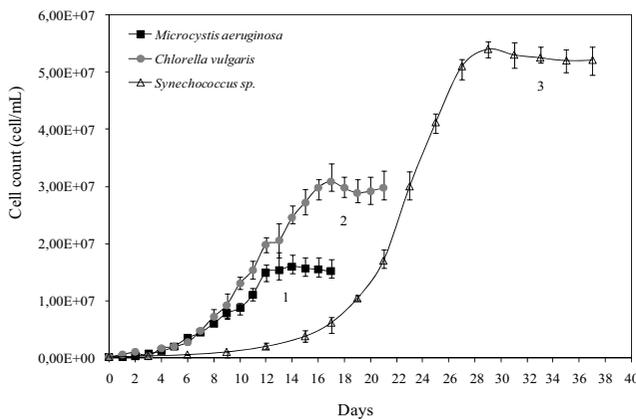


Fig. 1. The growth phases of *Microcystis aeruginosa*, *Chlorella vulgaris*, and *Synechococcus* sp. (1), (2) and (3) shows stationary phase of algae and cyanobacteria. $T = 25\text{ }^{\circ}\text{C}$, $\text{pH} = 9.0$. Error bars are \pm S.D. ($n = 3$).

2.2. Zeta potential measurements

The zeta potential measurements of cells were obtained using a Micro-Electrophoresis Apparatus Mk II (Rank Brothers, UK). The zeta potential was evaluated at a room temperature of $20 \pm 1\text{ }^{\circ}\text{C}$ and in suspension under an applied electric field of 80 mV. The zeta potential was calculated according to the Smoluchowski equation. The pH of algae and cyanobacteria suspension was adjusted from 2 to 10 using of HCl and NaOH to investigate the effect of pH on the zeta potential of

the cell surface. Furthermore, the zeta potential of the coagulated samples was measured as a function of the coagulant dosage after the coagulation experiments. For each species, triplicate cultures were taken for measurements and for each data set, approximately, 10 readings were taken. The average values are reported here.

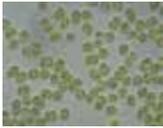
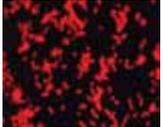
2.3. Jar test

At the early stationary phase, cell concentration for *Microcystis aeruginosa*, *Chlorella vulgaris* and *Synechococcus* sp. were $1.6 \times 10^7\text{ cell mL}^{-1}$, 3.1×10^7 and 5.4×10^7 , respectively. Since the population density was at its highest at this stage, the algae and cyanobacteria suspensions were diluted prior to the coagulation experiments to a concentration more often observed in surface water. Deionized water to which 0.35 mM NaHCO_3 (CAS 7542-12-3) had been added was used for this purpose. Approximately, 1.2×10^6 *Microcystis aeruginosa*, 1.3×10^6 *Chlorella vulgaris* and 1.5×10^6 *Synechococcus* sp. cells were used for the coagulation experiments.

Jar tests were performed in 1-l beakers using a six paddle jar test apparatus at a room temperature of $20 \pm 1\text{ }^{\circ}\text{C}$. A wide range of PACl (CAS 1327-41-9) dose (0–500 mg/l) during the experiment was used as coagulant to reach isoelectric point (IEP) of each species. The mixing velocity and time was designated based on the previous studies on algae removal [6,9]. The pH of the coagulation system was adjusted to pH 6.5 and 7 using the appropriate amounts of HCl and NaOH in

Table 1

Summary of cell characteristics in terms of average cell surface area where $r =$ radius ($n = 3$).

Species	Photo	Cell size diameter [18]	Surface area equation [16]	Cell surface area (μm^2)	Stationary phase concentration (cell mL^{-1})
<i>Microcystis aeruginosa</i>		5.5 μm	Sphere: $4\pi r^2$ where, $r = 2.75$	95	$1.6 \times 10^7 \pm 1.3 \times 10^6$
<i>Chlorella vulgaris</i>		4 μm	Sphere: $4\pi r^2$ where, $r = 2$	50	$3.1 \times 10^7 \pm 3.2 \times 10^6$
<i>Synechococcus</i> sp		2 μm	Sphere: $4\pi r^2$ where, $r = 1$	12	$5.4 \times 10^7 \pm 4.3 \times 10^6$

the slow mixing period. Previous studies showed that PACl provide highest coagulation efficiency in this pH range [22]. The initial rapid mixing was conducted at 150 rpm for 3 min followed by a slow mixing at 30 rpm for 30 min. The suspension was left to settle for 30 min. After settling, samples were taken from 2 cm below the surface for turbidity measurements. Turbidity was measured using a Water Analyzer WA 6000 Turbidimeter (Nippon Denshoku, Japan). The zeta potentials of the suspensions were also measured via a zeta analyzer (Rank Brothers, UK).

Jar tests experiments were repeated to investigate the relation between turbidity and algae removal. Algae and cyanobacteria samples were taken at the early stationary phase and diluted with tap water prior to coagulation experiments. As was mentioned earlier in the text, approximately, 1.2×10^6 *Microcystis aeruginosa*, 1.3×10^6 *Chlorella vulgaris* and 1.5×10^6 *Synechococcus* sp. cells were used in the coagulation experiments. The turbidity of the suspensions was measured before the experiments to define the turbidity equivalent of the concentrations of the species studied. The number of species showed a linear relationship with turbidity (Fig. 2).

3. Results and discussion

3.1. Effect of pH on the zeta potential of different algae systems

The zeta potential is an important parameter of double layer repulsion for individual organic and inorganic particles, and is commonly used in explanations of coagulation efficiency [13]. Since the most important factor affecting the zeta potential is pH, the influence of

pH on the zeta potential of a variety of cell species was examined. Fig. 3 shows the average zeta potential measurements for *Microcystis aeruginosa*, *Chlorella vulgaris* and *Synechococcus* sp. from three independently grown cultures at different pH sp. The zeta potential measurements reveal that each of the three species has a negative charge in the pH range typical of natural water. As the pH decreases from 7.0 to 6.0 the zeta potential of *Microcystis aeruginosa* increased sharply and moved from the negative side (-23.48 ± 2.2 mV) into the positive side (22.4 ± 1.5 mV). The IEP was determined to be approximately 6.3. At the IEP, the concentration of negatively charged surface sites is equal to the positively surface charge groups, most of which are thought to be amine groups. As the pH decreases from the IEP, the protonation of the base surface groups generates positive zeta potential at the cell surface [14]. For *Synechococcus* sp., the zeta potential decreased sharply from pH 6.0 to 5.0, and the IEP was achieved at approximately pH 5.5. The zeta potential at pH 6.0 and 5.0 was -8.49 ± 2.1 and 9.16 ± 3.1 mV for *Synechococcus* sp. Zeta potential measurements reveal that *Microcystis aeruginosa* and *Synechococcus* sp. exhibit a net positive surface charge below a pH value of approximately 6.3 and 5.5, respectively. For *Chlorella vulgaris*, the zeta potential measurements showed that the IEP of *Chlorella vulgaris*, at approximately 2.6 was lower than those of both *Microcystis aeruginosa* and *Synechococcus* sp., in contrast to other species, the zeta potential of *Chlorella vulgaris* decreased slowly from pH 10.0 to 2.0. Moreover, the zeta potential of *Chlorella vulgaris* was significantly less over a larger pH range than it was for both *Microcystis aeruginosa* and *Synechococcus* sp. The zeta potentials of each of the three species reached a maximum value at

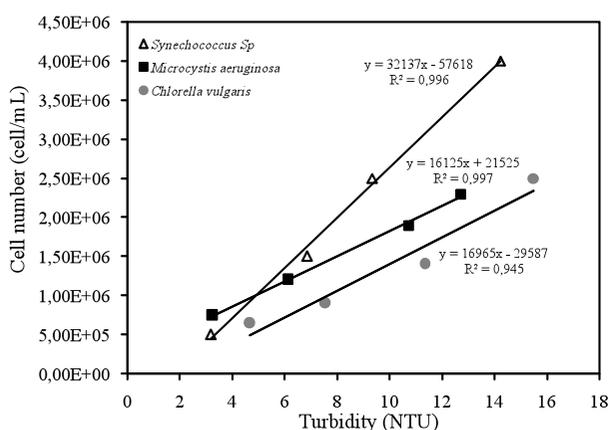


Fig. 2. The turbidity equivalent of the concentrations of the species studied.

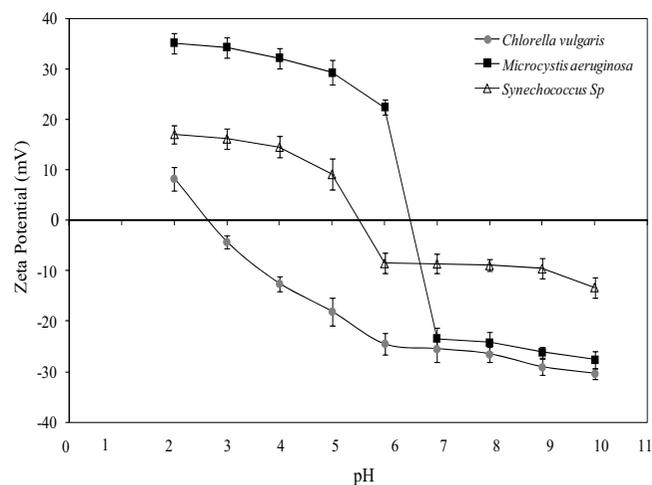


Fig. 3. Zeta potential of *Microcystis aeruginosa*, *Chlorella vulgaris* and *Synechococcus* sp as a function of pH. Error bars are \pm S.D. $T = 20 \pm 1$ °C ($n = 5$).

pH 2. The zeta potentials were +35.1 mV for *Microcystis aeruginosa*, +17.11 mV for *Synechococcus* sp. and +8.22 mV for *Chlorella vulgaris*. The pH was significantly more effective in neutralizing in the case of *Microcystis aeruginosa* than the other two species.

Each of the species had different sensitivities to changes in pH due to their different surface characteristics. When the pH is low, the neutralization of the charge of all species was more effective. It should be remembered; however, that pH of drinking water is required to be adjusted from 6.5 to 8.5 in accordance with the Environmental Protection Agency (EPA) [21]. Since drinking water treatment is generally conducted in this range, the coagulation experiments in this study were undertaken at both pH 6.5 and 7.0.

3.2. Coagulant demand and removal efficiency of cells

The zeta potential of each of the algae systems was monitored in the current study as a function of the coagulant dose and correlated with the cell removal rate. Fig. 4 shows that the zeta potential changes in all three systems

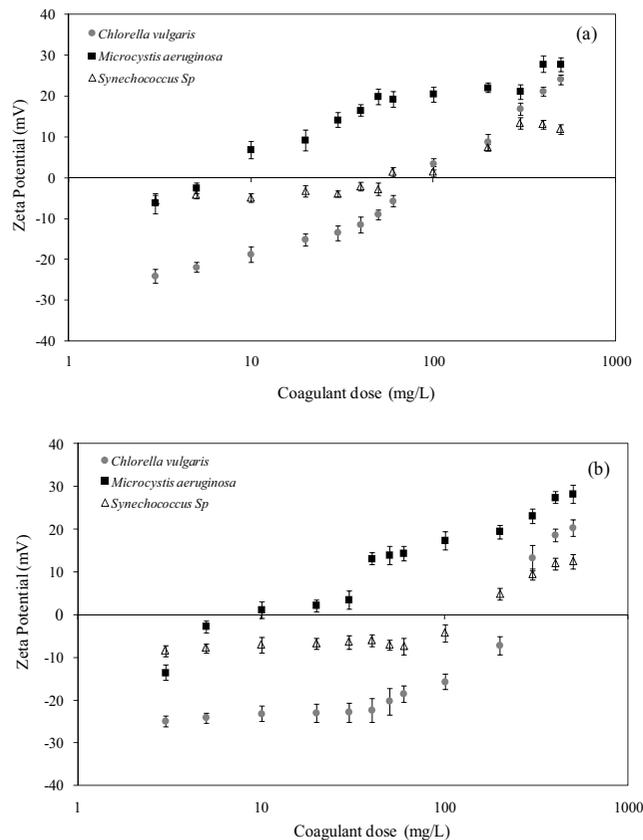


Fig. 4. Zeta potential of *Synechococcus* sp, *Microcystis aeruginosa* and *Chlorella vulgaris* as a function of coagulant dosage at pH 6.5 (a) and 7 (b). Error bars are \pm S.D. $T = 20 \pm 1$ °C. ($n = 5$).

with an increase in the coagulant dose at pH 6.5 and 7. The results indicated that the coagulant was significantly more effective at neutralizing the zeta potential of *Microcystis aeruginosa* than that of *Chlorella vulgaris* and *Synechococcus* sp. It is interesting that despite having a larger surface area than the other two species, *Microcystis aeruginosa* required a relatively low coagulant dose (pH 6.5 and 7.0) for charge neutralization. For *Microcystis aeruginosa*, the maximum removal efficiency was achieved at coagulant doses of 100 mg/L at pH 6.5 and 400 mg/L PACl at pH 7.0, achieving 98.9% and 95.1% cell removal, respectively. On the other hand, for *Chlorella vulgaris*, coagulant doses of 200 mg/L and 300 mg/L PACl were required to obtain 90.6% and 80.4% cell removal at pH 6.5 and 7.0, respectively. It is interesting that despite the smaller surface area of and less negative zeta potential of *Synechococcus* sp. than *Chlorella vulgaris*, a larger coagulant dose was required for the maximum removal. Coagulant doses of 300 mg/L at pH 6.5 and 500 mg/L at pH 7.0 required for maximum cell removal, at 55.7% and 32.4%, respectively. The low removal efficiency of *Synechococcus* sp. may be attributed to their low densities and size [19,20], which meant that the formed microflocs did not form a sediment. In other words, while the factors which govern the removal of the *Microcystis aeruginosa* and *Chlorella vulgaris* are the charge neutralization and sweep flocculation mechanism, in the case of *Synechococcus* sp., it is only the charge neutralization.

The results of this investigation differ somewhat to those reported in the literature. For instance, Henderson et al. [15] reported that the coagulant was significantly more effective at neutralizing the charge of *Chlorella vulgaris* than that of *Microcystis aeruginosa* at pH 7. The differences between results can be explained by the coagulation inhibition effect of the hydrophilic compounds on the cell surface. Recent studies have shown that negatively charged bacterial hydrophilic compounds with a molecular weight of more than 10 kDa have a significant inhibition effect on coagulation [16]. If stoichiometry is assumed to exist between neutralization and coagulant addition, the hydrophilic compounds must be significantly contributed to the coagulant demand. This has been demonstrated in a study of algogenic organic matter (AOM). AOM with a high affinity to metals in the coagulant induce a reduction in the coagulation efficiency due to the formation of complexes between metals and AOM which could bring about the excess consumption of coagulant [17]. It is likely that *Chlorella vulgaris* and *Synechococcus* sp. have significantly more AOM than *Microcystis aeruginosa*, which explains its higher coagulant consumption.

The zeta potential operation range for each species to obtain good removal was monitored at pH 6.5 and 7.0 (Fig. 5). The zeta potential operational ranges were

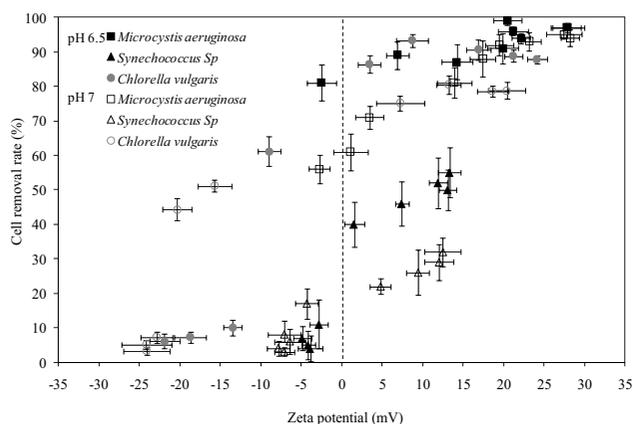


Fig. 5. Cell removal rate of *Microcystis aeruginosa*, *Chlorella vulgaris* and *Synechococcus* sp. at pH 6.5 and 7.0. Error bars are \pm S.D. $T = 20 \pm 1$ °C. ($n = 5$).

different for each system depending on the species and pH; for instance, at pH 6.5, *Microcystis aeruginosa* was successfully removed between -2.1 mV and $+20.45$ mV, whereas the zeta potential range for *Chlorella vulgaris* removal was much narrower, at $+3.45$ mV and $+8.71$ mV for optimum removal. *Synechococcus* sp. had a similar optimal range of $+7.41$ mV and $+13.33$ mV at pH 6.5, which was closer to the removal range for *Chlorella vulgaris* than that for *Microcystis aeruginosa*. These results were also compared with those obtained at pH 7.0. It was observed that much higher the zeta potential values were required at pH 7.0 for optimum cell removal than at pH 6.5. The optimal zeta potential range for *Microcystis aeruginosa*, *Chlorella vulgaris* and *Synechococcus* sp. removal were observed between $+13.88$ mV and $+23.14$ mV, $+7.2$ mV and $+13.2$ mV and $+9.4$ mV and $+12.46$ mV, respectively.

These results showed that coagulation experiments conducted at pH 6.5 achieved slightly more positive zeta potential. It was also noted that no decrease in the removal efficiency was observed at positive zeta potentials. Optimal cell removal was obtained irrespective of pH if the zeta potential was maintained between $+8.71$ mV and $+28.2$ mV.

3.3. The zeta potential operational range for turbidity removal

The relationship between the residual turbidity and zeta potential of various species were examined at pH 6.5. In each instance, the residual turbidity was decreased as the magnitude of zeta potential became lower. Fig. 6 illustrates the zeta potential operational ranges for each species which corresponding to the turbidity removal. The residual turbidity in the *Microcystis aeruginosa* system decreased 0.14 NTU as a function of

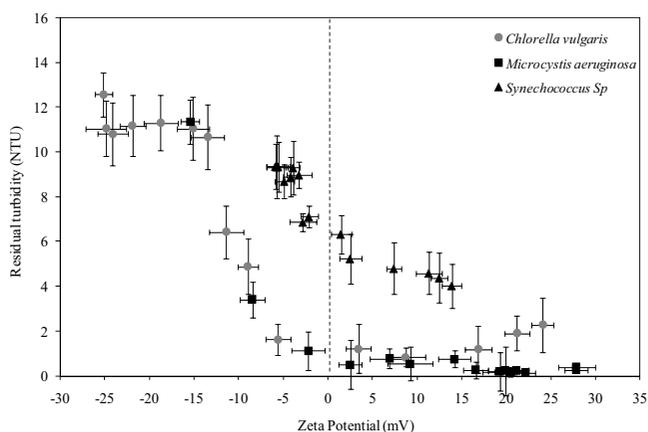


Fig. 6. Zeta potential and residual turbidity. Error bars are \pm S.D. $T = 20 \pm 1$ °C, at pH 6.5. ($n = 5$).

the coagulant dose. The corresponding zeta potential at that point was $+20.12 \pm 1.3$ mV with coagulant dose of 100 mg/l PACl for *Microcystis aeruginosa*. It was demonstrated that high zeta potential provides good removal for *Microcystis aeruginosa*. Moreover, high coagulant dose did not cause to restabilization of *Microcystis aeruginosa* system. In the previous studies, it is proposed that the absence of a restabilization in *Microcystis aeruginosa* system is attributable to large molecular weight protein and carbohydrates on the surface of *Microcystis aeruginosa* cell acting as polymer aids and overcoming repulsive electrostatic force [15]. In addition, it was observed that flocs formed in the *Microcystis aeruginosa* system were larger and formed more quickly than those in other systems. *Microcystis aeruginosa* removal is attributed to both the charge neutralization and sweep coagulation mechanisms. The flocs formed showed the higher sedimentation performance, and this allowed for a higher turbidity removal. There were three different zeta potential ranges observed for *Chlorella vulgaris*. The first range was the initial range of turbidity removal. At this range, however, very low turbidity removal was observed; the zeta potentials were between -25.16 ± 1.1 mV and -13.49 ± 1.8 mV. The second range was the maximum removal range of turbidity where *Chlorella vulgaris* removal is attributed charge neutralization mechanism [23]. The zeta potentials were between -11.4 ± 1.9 mV and $+8.71 \pm 2.2$ mV in this range. Residual turbidity at this range was 0.84 NTU with coagulant dose of 200 mg/l PACl. The third range was the restabilization range; at this range, residual turbidity increased to 2.28 NTU again where the zeta potential values ranged between $+16.87 \pm 1.5$ mV and $+24.1 \pm 1.2$ mV. Restabilization of *Chlorella vulgaris* system could be explained by high coagulant dose. This sequence of removal ranges is only observed for *Chlorella vulgaris* system, in contrast to other systems.

There was very low turbidity removal in the *Synechococcus* sp. system observed when the zeta potential range of the system was between -5.87 ± 1.2 mV and 0 mV. However, when the zeta potential of the *Synechococcus* sp. system moved from the negative side into the positive side, the turbidity removal increased. The zeta potential value which provided for the maximum turbidity removal was $+13.91 \pm 1.1$ mV with dose of 300 mg/l PACl for *Synechococcus* sp. system and the residual turbidity at that point was 4.02 NTU. More difficulty was encountered removing turbidity from the *Synechococcus* sp. system than from the *Microcystis aeruginosa* and *Chlorella vulgaris* systems. This may be due to the low density and small size [19] of the formed flocs, both of which would contribute to the low sedimentation performance of the *Synechococcus* sp. flocs. The results showed that charge neutralization is the most effective method for the removal of turbidity caused by *Synechococcus* sp. The results demonstrated that a positive zeta potential contributed to turbidity removal in each algal system.

The turbidity removal performance of mixtures of algae systems was also investigated in this study. Fig. 7 shows the removal performance results of three mixtures of algae systems at pH 6.5. The high turbidity removal observed in the *Microcystis aeruginosa* and *Chlorella vulgaris* mixture can be attributed to the high sedimentation performance of the very large flocs formed, which contributed to the turbidity removal. The lowest turbidity removal was observed in the ternary system including *Synechococcus* sp. which in contrast to the *Microcystis aeruginosa* and *Chlorella vulgaris* mixture, was characterized by much smaller floc formation. This is considered the reason for the low turbidity removal for the ternary system. The results showed that *Synechococcus* sp. removal is difficult than other species; which present a challenge for water

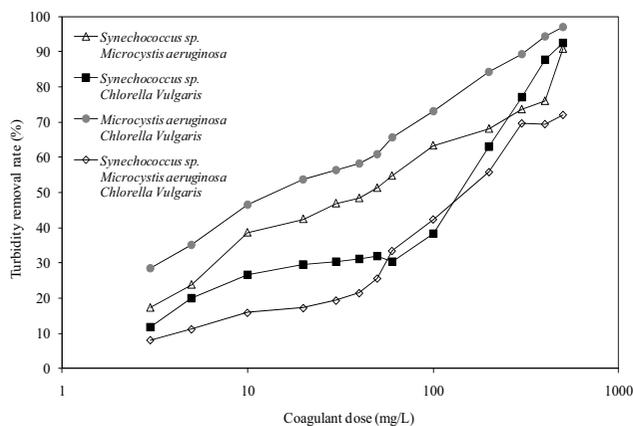


Fig. 7. Comparison of turbidity removal of mixture systems. $T = 20 \pm 1$ °C, pH = 6.5.

treatment process, however, the mixtures of algae systems contributed the removal of *Synechococcus* sp. It can be attributed to *Synechococcus* sp. stuck in massive floc formed by *Microcystis aeruginosa* or *Chlorella vulgaris*. Turbidity removal efficiencies were observed as 97% for the *Microcystis aeruginosa* and *Chlorella vulgaris* mixture, 92% for the *Synechococcus* sp. and *Chlorella vulgaris* mixture, 90% for *Synechococcus* sp. and *Microcystis aeruginosa* mixture and 72% for the mixture of three. It was clearly shown that the species of algae and cyanobacteria in the raw water has a significant effect on the coagulation efficiency making it important to determine the type of cells in the raw water for drinking water treatment.

4. Conclusions

The zeta potential and coagulation behaviors of different cyanobacteria and algae were investigated by measuring the zeta potential, taking cell counts and assessing the turbidity removal in various systems. Several coagulation tests were performed with different coagulant doses. The specific conclusions are as follows:

1. The charge of all species is more effectively neutralized at low pH values. The pH was significantly more effective at neutralizing the *Microcystis aeruginosa* systems than the other two species.
2. The coagulation process is affected by surface charge. At pH 6.5, the optimal zeta potential band were between -2.1 mV and $+20.45$ mV for *Microcystis aeruginosa*, $+3.45$ mV and $+8.71$ mV for *Chlorella vulgaris* and $+7.41$ mV and $+13.33$ mV for *Synechococcus* sp. at 20 ± 1 °C using 0–500 mg/l PACl. The optimal zeta potential range for *Microcystis aeruginosa*, *Chlorella vulgaris* and *Synechococcus* sp. removal is much narrower at pH 7 than at pH 6.5 under the same the experiment conditions. Positive zeta potential was shown contribute to turbidity removal in each algal system.
3. Maximum removal efficiency was at coagulant doses of 100 mg/L for *Microcystis aeruginosa*, 200 mg/L for *Chlorella vulgaris* and 300 mg/L PACl for *Synechococcus* sp. at pH 6.5, with cell removal rates of 98.9%, 90.6% and 55.7%, respectively.
4. More difficulty was encountered removing *Synechococcus* sp. than *Microcystis aeruginosa* and *Chlorella vulgaris*. This could be attributed to the low sedimentation performance of the *Synechococcus* sp. flocs formed due to their low density small size.
5. The particular species of algae and cyanobacteria in the raw water has a significant impact on the coagulation efficiency.

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