# Investigation of Coagulation Conditions for Enhanced Picophytoplantkon Removal in Drinking Water Treatment

| | |
|---|---|
| 著者 | TUGRUL SELAMI AKTAS |
| 号 | 57 |
| 学位授与機関 | Tohoku University |
| 学位授与番号 | 工博第4730号 |
| URL | http://hdl.handle.net/10097/62064 |

| | |
|---|---|
| 氏名, | タグルス　セラミ　アクタス<br>TUGRUL　SELAMI　AKTAS |
| 授与学位, | 博士（工学） |
| 学位授与年月日, | 平成２４年９月２５日 |
| 学位授与の根拠法規 | 学位規則第４条第１項 |
| 研究科，専攻の名称 | 東北大学大学院工学研究科（博士課程）土木工学専攻 |
| 学位論文題目, | Investigation of Coagulation Conditions for Enhanced Picophytoplankton Removal in Drinking Water Treatment（浄水処理におけるピコ植物プランクトン除去能強化のための凝集条件） |
| 指　導　教　員 | 東北大学教授　西村　修 |
| 論文審査委員, | 主査　東北大学教授　西村　修　　東北大学教授　大村　達夫<br>東北大学教授　原田　秀樹 |

# 論文内容要旨,

Picophytoplankton (picoplanktonic cyanobacteria) is a small plankton ranging between 0.2 and 2 μm in size, comprised of picocyanobacteria and eukaryotic phototrophs. The presence of picophytoplankton and its metabolites in drinking water sources can cause a series problems for drinking water treatment. For instance, picophytoplankton cells caused high turbid water problem of treated water and filter clogging problem in rapid sand filtration process. Their intracellular metabolites also contributed to the production of undesired tastes and odors, the formation of assimilable organic carbon (AOC), disinfection byproducts (DBPs) and cyanotoxins. Therefore, picophytoplankton has recently become one of the most important targets in drinking water treatment.

Despite the many negative effects of picophytoplankton, the removal of picophytoplankton from drinking water has not been well studied. Therefore, the studies need to be done to enhance coagulation process for the effective removal of picophytoplankton from drinking water. In this study, the coagulation mechanisms of picophytoplankton and how to improve picophytoplankton removal with coagulation process were discussed based on the results of coagulation experiments under various physical and chemical conditions, e.g. coagulant type and rapid mixing speed / time.

Experiments were conducted using both raw water and artificial water samples containing picophytoplankton. In this study, as a first step, the optimal pH conditions and effective coagulant type for picophytoplanton removal were investigated. The results showed that the optimal pH was ranged from 5.5 to 6.5 for the removal of picophytoplankton and turbidity in coagulation process. In addition, poly silica iron (PSI) showed the best removal performance in comparison to poly aluminum chloride (PAC), ferric chloride ($FeCl_3$) and aluminum chloride (Alum).

In this study, the zeta potential of picophytoplankton formed flocs was investigated to make it clear the coagulation mechanism of different algal systems such as *Microcystis aeruginosa*, *Chlorella vulgaris* and *Synechococcus* sp. These species are commonly found in water sources and associated with water quality and treatment problems in water treatment plants. As it well known that the stability of the system depend on the zeta potential of the system. 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. 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 contributes to turbidity removal of the system. As such, the zeta potential is an important parameter when investigating the coagulation mechanism. From the result of *Microcystis aeruginosa* system, the lowest residual turbidity of 0.14 NTU was obtained under the conditions of zeta potential of +20.12 ± 1.3 mV and coagulant dose of 100 mg/l PACl. Moreover, high coagulant dose (over 100 mg/L) didn't cause to restabilization of *Microcystis aeruginosa* system. It was demonstrated that high zeta potential provides good removal for *Microcystis aeruginosa.* In addition, it was observed that flocs formed in the *Microcystis aeruginosa* system were larger and formed more quickly than those in other systems.

In *Chlorella vulgaris* system, the relationship between coagulant dosage and residual turbidity was divided into three parts, which had different zeta potential ranges. At first part with relatively low coagulant dosage, very low turbidity removal was observed and the zeta potentials were between -25.16 ± 1.1 and -13.49 ± 1.8 mV. In the second part the maximum removal of turbidity occurred and the zeta potentials were between -11.4 ± 1.9 and +8.71 ± 2.2 mV. In the third part the residual turbidity increased with the increase of coagulant dosage and the zeta potentials were between +16.87 ± 1.5 mV and +24.1 ± 1.2 mV. The result indicated the restabilization of flocs in *Chlorella vulgaris* system occurred under high coagulant dosage. Restabilization was only observed for *Chlorella vulgaris* system, in contrast to other systems.

The very low turbidity removal in *Synechococcus* sp. system was observed, when the zeta potential range of the system was between -5.87 ± 1.2 and 0 mV. However, when the zeta potential of the *Synechococcus* sp. system moved from the negative side into the positive side by addition of coagulant, the turbidity removal increased. The maximum turbidity removal was achieved when the zeta potential value was +13.91 ± 1.1 mV with coagulant dosage of 300 mg/l PACl. Comparing with three systems, removal of turbidity was most difficult in the *Synechococcus* sp. system than from the *Microcystis aeruginosa* and *Chlorella vulgaris* systems. This may be due to the lower density and smaller size of the formed flocs in *Synechococcus* sp. system. The results also showed that interparticle bringing was the most effective method for the removal of turbidity caused

*Synechococcus* sp. The results demonstrated that a positive zeta potential contributed to turbidity removal in each algal system. Zeta potential experiments revealed that optimal cell removal was obtained irrespective of pH if the zeta potential was maintained between +8.71 mV and +28.2 mV. In the literature, the optimum zeta potential range has been reported to be between -14.5 mV and +12 mV for algae (*Chlorealla vulgaris*). In our case, we investigated that more species and suggested that narrower zeta potential range for optimum cell removal.

The effect of different velocity gradients and rapid mixing durations on floc growth in the cyanobacterial system were investigated to optimize and improve the mixing conditions in coagulation-flocculation process. It was noted that the floc size of both the *Synechococcus* sp. and kaolin system first increased and then decreased with increased *G* in the coagulation process, which demonstrated that there were three different ranges for both *Synechococcus* sp. and kaolin flocs formation: the low velocity gradient range ($G < 200\ s^{-1}$), which was the aggregation-dominated range; the mean velocity gradient range ($G$= 250 $s^{-1}$ for *Synechococcus* sp.-PSI flocs and $G$=546 $s^{-1}$ for kaolin-PSI flocs; $G$= 200 $s^{-1}$ for *Synechococcus* sp.-PAC flocs and $G$=390 $s^{-1}$ for kaolin-PAC flocs), where coagulation rates were maximized and breakup was minimal; and the high velocity gradient range ($G > 250\ s^{-1}$ for *Synechococcus* sp.-PSI flocs and $G > 546\ s^{-1}$ for kaolin-PSI flocs; $G > 200$ *Synechococcus* sp.- PAC flocs and $G > 390\ s^{-1}$ for kaolin-PAC flocs), where flocs breakup was dominant. In the aggregation-dominated range, the aggregate size increased in proportion to the share rate, but resulted in smaller flocs due to the low particle collision rates. Although breakup most likely occurred, the breakage was considered to have relatively insignificant impact on the overall aggregate sizes. In the mean velocity gradient range, a balance between the rate of aggregation and the rate of breakage was reached; the flocs reached their maximum size. In the breakup dominant range, collisions between the flocs produced more, but smaller flocs. In the beginning of this range, while a slight decrease was noted in the size of *Synechococcus* sp. flocs, the size of the flocs in the kaolin system significantly decreased, indicating less breakup in the *Synechococcus* sp. flocs in comparison to kaolin flocs. At the end of this range, the re-growth ability of *Synechococcus* sp. flocs was observed to be higher than the kaolin flocs. The physical parameters experiments indicated that at mean *G* value flocs formation was maximized, producing the largest flocs. In addition, in contrast to previous studies, relatively high velocity gradients are suggested for the coagulation process in drinking water treatment. Consequently, if the above mentioned coagulation conditions are provided during the experiments picophytoplankton removal efficiency can be increased in drinking water treatment plants.

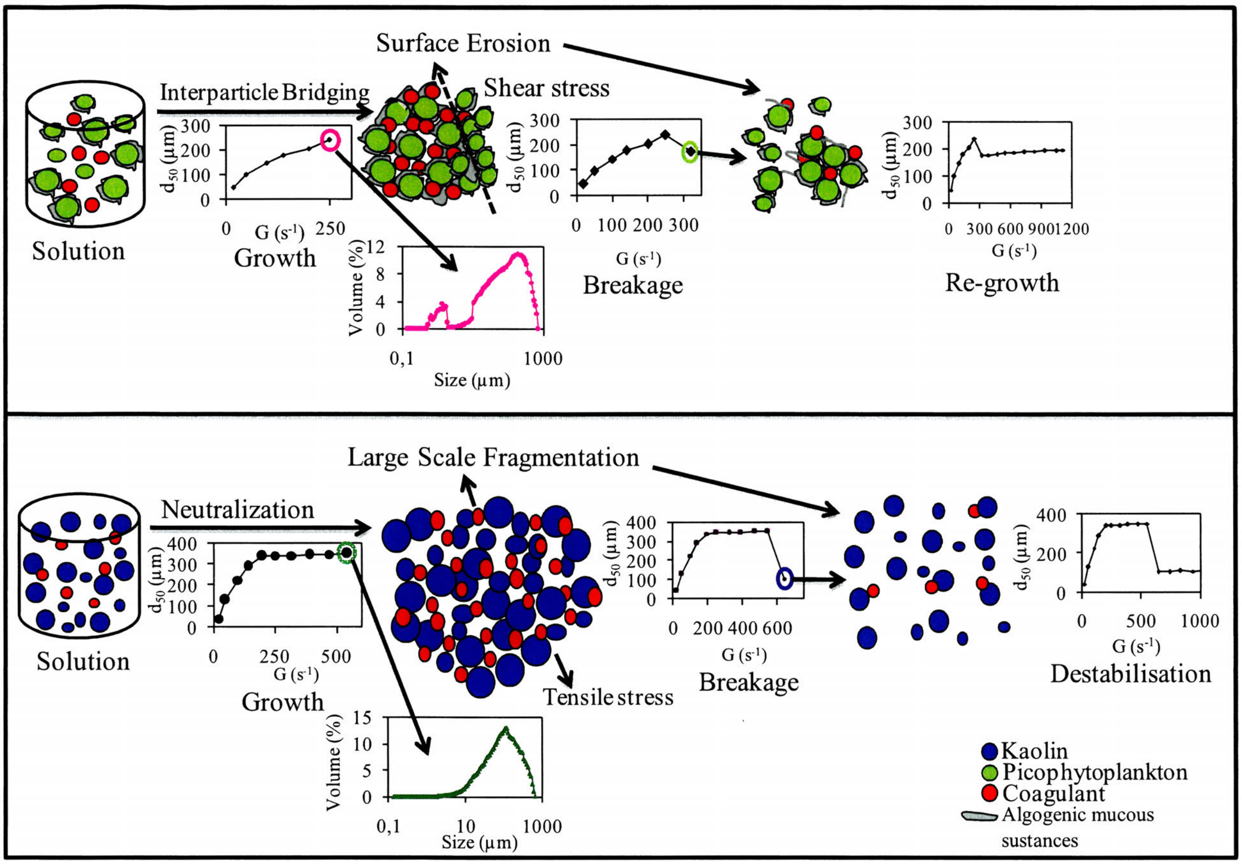


**Figure 1.** The mechanism of flocs formation, breakage and re-growth

As a result, this study showed that PSI the best coagulant type for picophytoplankton removal and can be recommended in drinking water treatment. In addition, if the zeta potential of picophytoplankton system be keep over +7 mV, the optimum picophytoplankton removal can be provided. Lastly, for picophytoplankton system, the optimal rapid mixing conditions can be suggested that *G* value is 200 $s^{-1}$ for PSI and the rapid mixing time is 90 s.

# 論文審査結果の要旨

水道原水に混入するピコ植物プランクトンは，凝集やろ過などの浄水プロセスで深刻な障害を発生させるため，効果的な除去技術の開発が求められている．本論文は，水道原水に混入するピコ植物プランクトンの凝集処理のために，ピコ植物プランクトンの混入する実原水，およびピコ植物プランクトン *Synechococcus* sp.を培養して作成したモデル原水を用いて凝集特性を検討したもので，全編 6 章よりなる．

第 1 章「緒論」では，本研究の背景としてピコ植物プランクトンによる浄水障害解決の必要性を述べ，本研究の目的および意義についてまとめた．

第 2 章「既往研究の整理」では，ピコ植物プランクトンによる浄水障害，および植物プランクトンの凝集特性に関する既往の研究を整理し，ピコ植物プランクトンの凝集特性に関して検討すべき研究課題をまとめた．

第 3 章「ピコ植物プランクトンの凝集に及ぼす凝集剤の影響」では，ピコ植物プランクトンの凝集に及ぼす pH と凝集剤種類（塩化アルミニウム，塩化第二鉄，ポリ塩化アルミニウム，ポリシリカ鉄），および添加量の影響を検討し，実原水では pH6.5 で凝集剤種類にかかわらず最大の除去効果が得られること，ポリシリカ鉄により残留濁度が最も小さくなり，これに要する凝集剤注入量は最も少ないことを明らかにした．これらは有用な知見である．

第 4 章「ピコ植物プランクトンおよび植物プランクトンの凝集特性のゼータポテンシャルによる比較」では，凝集過程におけるピコ植物プランクトンの作るフロックの表面荷電の変化を植物プランクトンと比較しながら評価した．*Chlorella vulgaris* および *Microcystis aeruginosa* はゼータポテンシャルが増加し，荷電中和の生じる条件で急激に残留濁度を低下させるのに対し，*Synechococcus* sp.では荷電中和が生じても残留濁度の急激な低下は起こらず，凝集剤の添加量の増加とともに徐々に残留濁度の低下が生じることがわかった．これらは新規な知見である．

第 5 章「植物ピコプランクトンの凝集における攪拌条件の影響」では，ピコ植物プランクトンの凝集のための最適な攪拌条件に関する実験的検討を行った．比較のため用いた粘土粒子カオリンの最適 G 値の範囲が 200 から 400$sec^{-1}$と広いのに対して *Synechococcus* sp.は G 値の増加とともにフロック径を急激に増大させ，最適な G 値 250$sec^{-1}$を過ぎると急激にフロックが分散すること，また攪拌時間にも最適値が存在し 90sec であることが明らかになった．この理由はピコ植物プランクトンの作るフロックの強度が弱いためであり，ピコ植物プランクトンの凝集には比較的低 G 値で架橋作用を働かせることが有効であることが考えられた．これは特に新規かつ有用な知見である．

第 6 章「総括および展望」では，本研究の結論および今後の研究課題について整理した．

以上要するに本論文は，ピコ植物プランクトンの凝集特性として荷電中和よりも架橋作用によるフロック成長が重要であること，および除去能強化のための凝集剤種類としてポリシリカ鉄，攪拌条件として 250$sec^{-1}$で攪拌時間 90sec が最適であることを明らかにし，さらに従来指標であるゼータポテンシャルの植物ピコプランクトンの凝集に対する有用性と限界をまとめたもので，環境工学の発展に寄与することが少なくない．

よって，本論文は博士(工学)の学位論文として合格と認める．