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The use of alum as coagulant for removing cyanobacterial cells in drinking water

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ABSTRACT

A variety of problems can occur due to the presence of cyanobacteria in water resources used for drinking, agricultural, industrial, commercial, and recreational purposes. In addition, certain cyanobacteria genera are producers of several potent toxins, which endanger the human and animal health. Coagulation is the key step in water treatment process for algae and cyanobacteria, and their associated metabolites removal. The objective of this study was to examine the coagulation processes to optimize the removal of cyanobacterial cells from drinking water under various aluminum sulfate dose and pH values. The influence of cationic polyelectrolyte as a coagulant aid on the cells in accompany with aluminum sulfate was also studied. A set of jar test experiments at 200 rpm of rapid mixing, and 30 rpm of slow mixing and 30 min settling time were conducted to find the optimum chemical dose and pH. From the results of the tests, the optimum dose and pH for aluminum sulfate coagulant and polyelectrolyte were obtained corresponding to the lowest concentrations of cyanobacterial cells and turbidity.

Keywords: Eutrophication; Cyanobacteria; Microcystin; Coagulation; Aluminum sulfate

1. Introduction

Eutrophication is the enhancement of the natural process of biological production caused by nutrient enrichment, mainly nitrates and phosphates [1]. Some lakes are naturally eutrophic, however many water bodies are greatly accelerated by human activities resulting from municipal wastewater discharge or runoff from agricultural land [2]. This has led to a widespread of algae and cyanobacteria in fresh water, and thus has had a considerable impact on water resources used for drinking, agricultural, industrial, commercial, and recreational purposes.

Cyanobacteria, also known as blue-green algae, is often associated with eutrophic water [3]. Specific cyanobacterial species that cause algae blooming can release algal organic matters (AOM), consisting of cells, extracellular organic matters (EOM), and intracellular organic matters (IOM), into water during cell growth and lysis [4]. These organic matter entering downstream drinking water treatment systems can

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cause a variety of problems [2,5,6]. Various unpleasant taste and odor problems can occur [7]. Excessive cyanobacterial blooms can clog filters, and increase coagulant demand and formation of disinfection byproduct [8–11]. These lower the perceived quality of treated water and increase maintenance cost.

In addition, certain cyanobacteria genera such as *Anabaena*, *Aphanizomenon*, *Microcystis*, and *Pseudanabaena*, are producers of several potent toxins [12] called cyanotoxins, which endanger the human and animal health [13]. Contact with or ingestion of water containing cyanobacterial cells or toxins produce a variety of symptoms in humans including fever, headaches, muscle and joint pain, blisters, stomach cramps, diarrhea, vomiting, mouth ulcers, and allergic reactions. In some cases, seizures, liver failure, respiratory arrest, and (rarely) death may occur [14]. The hepatotoxic microcystins (MCs) produced by *Microcystis aeruginosa* are the most common cyanobacterial toxins found in water [3,7]. Microcystin-LR (MC-LR) is known to be one of the most toxic cyanotoxins in the water resources. The World Health Organization (WHO) and several countries have established guideline values ranging from 1 to 1.5 μg/L for MC-LR in drinking water [15]. Furthermore, the United States Environmental Protection Agency (USEPA) has placed MCs on the Drinking Water Contaminants Candidate List 3 [16].

In general, MCs are generally contained in cyanobacterial cells (intracellular toxins), unless the cell membrane is damaged or lysed (dissolved or extracellular toxins) [17]. The extracellular toxins are released from the cells and they are not so readily removed by the coagulation process as the intact cells are [18]. Thus, it is crucial to develop a method to remove cyanobacterial cells without causing cell lysis. It has been concluded that coagulation is the key step in water treatment process for algae and cyanobacteria, and their associated metabolites removal [19]. Coagulation involves the addition of chemicals to neutralize negative charges of colloids and prevents electrostatic repulsion between particles. Therefore, colloids tend to agglomerate and form flocs that are subsequently removed by sedimentation [15]. Since cyanobacterial cells possess negative surface charge, the surface charge can be neutralized by introducing coagulants. These coagulants can easily flocculate algal cells and form flocs [20,21].

Various researchers have investigated the studies on the coagulation process for the removal of cyanobacterial cells and cyanotoxins release. For example, Chow at al. [22] evaluated the effect of coagulation using aluminum sulfate (alum) on the integrity of cells of toxic *M. aeruginosa*. The results indicated that alum did not appear to cause lysis of cells of cultured *M. aeruginosa* nor increase the amount of MC-LR in the water. Sun et al. [23] showed that all cells were removed without damage to membrane integrity under the optimum coagulation conditions. They found that the $AlCI_3$ dose and shear did not cause the lysis of *M. aeruginosa* cells and release of MCs, but when the flocs were stacked over 6 d, the cells lysed and the MCs concentration increased above the background level. Han et al. [24] also investigated the effect of alum treatment on toxic *Microcystis* cells using a microcosm experiment designed to simulate common lakes or reservoirs. They reported that alum treatment caused cell damage and subsequent release of large amounts of MC-LR. Sun et al. [25] assessed the damage of polyaluminum chloride (PACl) coagulation/flocculation process on *M. aeruginosa* cells. Effects of coagulant dose, coagulation stirring, and floc storage time were comprehensively evaluated to regulate *M. aeruginosa* cell lysis and MCs release. Results showed that all cells were removed intact by the surface charge neutralization with PACl in the coagulation process. While in floc storage process, PACl caused obvious damage to cells and led to a large amount of MCs release. Li et al. [18] evaluated the effect of ferric chloride ($FeCl_3$) coagulation and the floc storage process on the integrity of *M. aeruginosa* cells and the intracellular MCs release in both the processes. The coagulant dose and mechanical actions caused no cell damage, and all the cells remained intact. Furthermore, 100-mg/L $FeCl_3$ was effective in removing the extracellular MCs.

Release of toxins by coagulation has not been reported in some studies, while release of toxins by coagulation has been reported in others. This indicated that efficiency of coagulation process can be strongly affected by the type and dose of coagulant, pH values, applied shear, and characteristics of the raw water such as turbidity, temperature, alkalinity, amount, and properties of colloids and suspended solids in water. If the coagulation operating conditions are not optimized, it is possible that the treatment process may cause cell damage and release toxins [26]. Only a few researchers have investigated the studies on the coagulation process using alum. Other types of coagulants have been studied for this application. And also, most of these have concentrated on the integrity of cells and floc storage time. There is slight information concerning characteristics of water, the optimum coagulant dose, and influence of pH and coagulant aid on the cells in company with coagulant. Although the characteristics of raw water, coagulant dose, and pH will vary system to system, the objective of this study was to examine the coagulation processes to

optimize the removal of cyanobacterial cells from drinking water under various doses and pH values. The evaluation has been carried out through bench-scale jar test for the most commonly applied coagulant of alum using natural water samples taken from Büyükçekmece Lake, which is a drinking water source, Istanbul, Turkey. The influence of cationic polyelectrolyte as a coagulant aid on the cells in company with alum was also studied. From the results of the tests, the optimum dose and pH for alum coagulant and polyelectrolyte were obtained corresponding to the lowest concentrations of cyanobacterial cells and turbidity. The results of this study can be used to make optimum process choices that reduce the risk of cyanobacterial cells in produced water.

2. Materials and methods

2.1. Materials

2.1.1. Cyanobacterial culture

The freshwater cyanobacteria culture *M. aeruginosa* was obtained from Algae Culture Collection of Istanbul University, Faculty of Fisheries, Department of Freshwater Biology, İstanbul, Turkey and cultured in BG11 medium. The cultures were grown in a light/-dark cycle (12/12) and at constant temperature (25˚C). Mixing was undertaken daily by hand.

2.1.2. Water

In the experimental system, cyanobacterial cultures in synthetic medium were transferred to natural water, which was taken from Büyükçekmece Lake (a drinking water source, Istanbul, Turkey). The characteristics of raw water qualities are summarized in Table 1. These analyses were done everyday during May–October 2013.

Table 1
Characteristics of Büyükçekmece lake water qualities

Parameter	Unit	Raw water	Average
pH		7.94–8.16	8.07
Temperature	(˚C)	15.00–25.78	22.46
Turbidity	(NTU)	1.73–36.20	5.70
Color	(Pt-Co)	12.50–50.00	15.74
Conductivity	(μS/cm)	551–573	563
Total hardness	(mg $CaCO_3$/L)	169–209	185
Alkalinity	(mg $CaCO_3$/L)	102–156	121
UV_{254}	(cm^{-1})	0.052–0.074	0.058

2.1.3. Chemicals

Alum ($Al_2(SO_4)_3{\cdot}18H_2O$; Sigma–Aldrich) was used as coagulant since this is the salt which is most commonly used in practice. One percentage of alum stock solution was prepared by dissolving 1-g alum to 100 mL of distilled water. Coagulants were added from a range of doses (0–100 mg/L) to determine the optimum coagulation conditions. pH was adjusted and varied between 5 and 8 units, i.e. at 5, 6, 7, 7.5, and 8 by adding 0.1 M NaOH and 0.1 M HCl. Polyelectrolyte stock solution was prepared as a 0.1% stock solution and added to the beaker in the flocculation period.

2.2. Methods

2.2.1. Intracellular toxin (Intra-MC)

For the extraction of intra-MC, samples were filtered through a glass fiber filter and stayed for 16–24 h in the lyophilizer. Lyophilized filters were extracted in 75% (v/v) aqueous methanol in an ultrasonic bath for 15 min. After this period, it was shaken in an orbital shaker at 100 rpm for 30 min and then centrifuged at 10,000× *g* for 10 min at room temperature. The supernatant was transferred to a vial, either analyzed immediately on HPLC-PDA or remained in the freezer (−18˚C, in the dark) until the analysis.

After extraction, samples were analyzed by high-performance liquid chromatography (HPLC) for intra-MC. Shimadzu HPLC-PDA system was used, which includes a high-pressure gradient pump (LC-20AT), an autosampler (SIL-20A), a column oven (CTO-10AS), and a photodiode array UV detector (SPD-M20A). A C18 column was used (Agilent Technologies-Nucleosil 100-5 C18 150 × 4.6M). The mobile phase used both a gradient of milli-Q water and acetonitrile with 0.05% (v/v) of trifluoroacetic acid. Microcystins were detected at 238 nm and chromatograms were analyzed between 200 and 300 nm compared to that of standard MC-LR, and expressed as intra-MC.

2.2.2. Coagulation experiments

Coagulation experiments were performed at room temperature (20 ± 2) in a standard jar test apparatus (Velp Scientifica FC6S) and equipped with six beakers of 1 L volume. During the experiments, pH was recorded and samples were collected for turbidity and intra-MC analysis. The jar tests were conducted in three series. The first series focused on intra-MC removal at all the different alum doses. The second

series focused on the effects of the same alum doses and different pH values. The third series focused on the effect of the same alum doses and pH, and different polyelectrolyte dose. A series of jar test experiments were rapidly mixed at 200 rpm for 2 min, and then slowly mixed at 30 rpm for 20 min, and at last the samples were allowed to settle for 30 min. At the end of settling period, the final turbidity was recorded and the samples were taken from the supernatant and filtered through a 0.45-μm pore size membrane to determine the optimum chemical dose and pH for intra-MC removal.

3. Results and discussion

A variety of water quality parameters can affect coagulation efficiency of coagulants, of which the dose of coagulant and pH are the two most important ones. Hence, the coagulation performance of alum was studied at different doses and pH. The results of turbidity and intra-MC removal with various alum doses are presented in Table 2. During the period of the experiments, the synthetic water samples had the following quality parameters: pH 8.15, temperature = 24.90˚C, turbidity = 9.00 NTU, color = 15.00 Pt-Co, conductivity = 561 μS/cm, alkalinity = 131 mg$CaCO_3$/L, and concentration of intra-MC = 10 μg/L.

Experiments were performed at natural pH of water without pH adjustment. The turbidity and intra-MC removal firstly increased with increasing alum dose from 20 to 60 mg/L. On increase in the dose from 80 to 100 mg/L, there was a slight decrease in the turbidity and intra-MC removal. The optimum coagulant dose in terms of turbidity and intracellular toxin removal were therefore determined as 60 mg/L and pH 7.16. Coagulation with alum at their optimum dose reduced the water sample's pH. This optimized dose was used in all coagulation experiments which were aimed at describing the influence of pH and polyelectrolyte dose. Alum with dose of 60 mg/L decreased the turbidity level to 0.62 NTU and intra-MC concentration to 4.11 μg/L. Though alum dose of 60 mg/l could remove turbidity, to achieve the maximum allowable turbidity by WHO guidelines [27], a significant amount of intra-MC still remained in the treated water. No further significant intra-MC removal was achieved after a dose of 60 mg/L. Similarly, Chow et al. [22] used about 60-mg/L alum to remove cyanobacterial cells from water. Chen et al. [5] examined and compared the effect of ozone and permanganate peroxidation on algae removal by alum coagulation. Without peroxidation, the algae removal was 84% at a 70 mg/L alum dose. Huang and Yeh [28] also examined how ozone and permanganate preoxidation affect coagulation of green algae (*Chodatella* sp.) and diatom (*Navícula* sp.) by alum. Without preoxidation, the residual algae concentration dropped to 50% at alum dose of 40 mg/L. Other types of coagulants have been studied by Sun et al. [23,25]. They found that the optimum coagulation conditions for the effective removal of cyanobacterial cells are a coagulant dose of 15 mg/L ($AlCl_3$) and 4 mg/L PACI.

Fig. 1 shows the turbidity and intra-MC removal efficiencies at different alum doses. The removal efficiencies were increased by the alum concentration from 20 to 60 mg/L. Turbidity removal varied between 91.8 and 89.1%, while intracellular toxin removal varied between 48.8 and 45.0%. The maximum removal efficiency of turbidity and intra-MC was 93.1 and 58.9%, respectively. Results indicated that removal efficiency was varied by alum dose.

The effect of pH for turbidity and intra-MC removal is shown in Table 3. Coagulation experiments with synthetic water were performed with an optimized dose of alum under various pH conditions. The turbidity and intra-MC removal firstly increased with increasing pH from 5 to 7. On increase in the pH from 7.5 to 8, there was a slight decrease in the turbidity and intra-MC removal. It was observed that the optimum pH for removal of turbidity and intra-MC by alum is approximately at pH 7.

Table 2
The results of turbidity and intra-MC removal with various alum doses

		Turbidity removal		Intra-MC removal	
Alum dose (mg/L)	Coagulation pH	NTU	%	μg/L	%
20	7.61	0.74	91.8	5.12	48.8
40	7.32	0.65	92.8	4.96	50.4
60	7.16	0.62	93.1	4.11	58.9
80	7.00	0.64	92.9	4.85	51.5
100	6.87	0.98	89.1	5.50	45.0

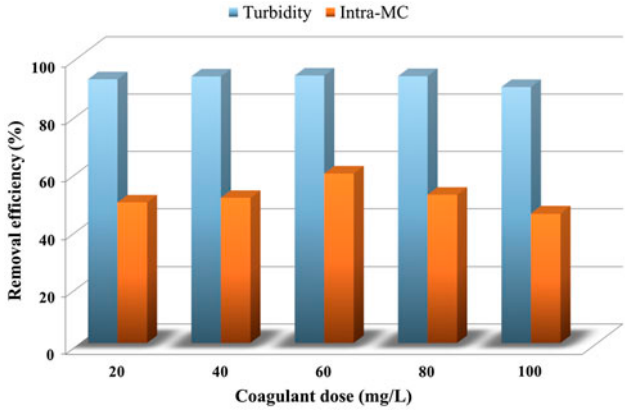

Fig. 1. Removal efficiency of turbidity and intra-MC with different alum doses.

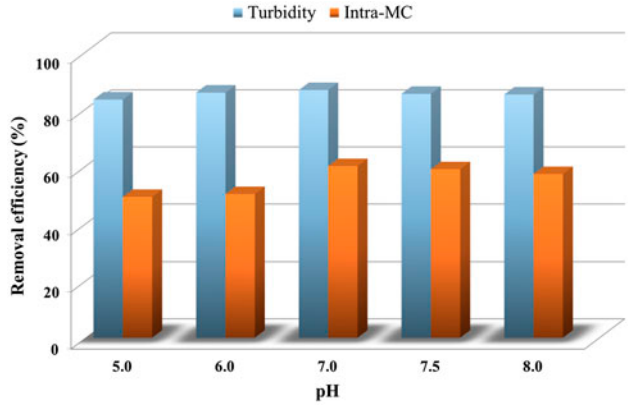

Fig. 2. Removal efficiency of turbidity and intra-MC with optimum alum dose and different pH.

Fig. 2 shows the coagulation removal efficiency of turbidity and intra-MC at different pH with 60 mg/L alum dose. As indicated in Fig. 2, removal efficiency increased with pH when pH could reach about 86.6 and 59.9% for turbidity and intra-MC, respectively. When the pH is between 7.5 and 8.0, the removal efficiency of turbidity and intra-MC slightly decreased.

Coagulation experiments were performed with an optimized dose of alum and pH under various polyelectrolyte doses to examine the additional benefits of adding polyelectrolyte in terms of treated water turbidity and intra-MC removal. The results are summarized in Table 4.

When polyelectrolyte was used as coagulation aid to alum, the cell removal of microcystin was slightly improved. Polyelectrolyte dose of 0.2 mg/L was enough to achieve turbidity and intra-MC removal. With greater polyelectrolyte dose, the turbidity and intra-MC removal decreased with increasing polyelectrolyte dose. At an alum dose of 60 mg/L, polyelectrolyte doses higher than 0.2 mg/L does not seem to be helpful to subsequent intra-MC removal via alum coagulation. In contrast, the optimum polymer dosage was found to be 0.75 mg/L by Huang and Yeh [28].

Fig. 3 illustrates the effect of different polyelectrolyte doses on turbidity and intra-MC removal at optimum alum dosage and pH value. While the turbidity removal in polyelectrolyte dose of 0.2 mg/L is 85.1%, intra-MC is 65.2%.

Fig. 4 presents the effects of pH and polyelectrolyte doses on *M. aeruginosa* cell removal. It can be seen that both pH and polyelectrolyte can slightly improve the cell removal through coagulation. Without pH change and polyelectrolyte, the cell removal was 59.9% at a 60-mg/L alum dose. The pH 7 was found to be ideal for higher performance of alum. With the pH change, the removal efficiency increased to 59.9% at the same alum dose. The removal efficiency for alum coagulation was improved by 1% combining pH change. When the 0.2 mg/L of polyelectrolyte dose was added, the removal efficiency increased to 65.2% at the same alum dose. The removal efficiency for alum

Table 3
The results of turbidity and intra-MC removal with optimum alum dose and different pH values

		Turbidity removal		Intra-MC removal	
Optimum alum dose (mg/L)	Coagulation pH	NTU	%	μg/L	%
60	5.00	1.51	83.2	5.09	49.1
60	6.00	1.29	85.7	4.99	50.1
60	7.00	1.21	86.6	4.01	59.9
60	7.50	1.33	85.2	4.12	58.8
60	8.00	1.35	85.0	4.29	57.1

Table 4
The results of turbidity and intra-MC removal with optimum alum dose and pH values with different polyelectrolyte doses

Optimum alum dose (mg/L)	Optimum pH	Polyelectrolyte dose (mg/L)	Turbidity removal		Intra-MC removal	
			NTU	%	μg/L	%
60	7.00	0.20	1.34	85.1	3.48	65.2
60	7.00	0.40	1.53	83.0	3.95	60.5
60	7.00	0.60	1.45	83.9	4.43	55.7
60	7.00	0.80	1.53	83.0	4.99	50.1
60	7.00	1.00	1.59	82.3	5.09	49.1

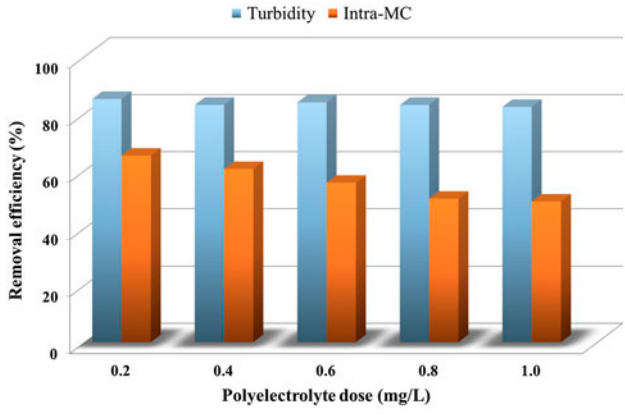

Fig. 3. Removal efficiency of turbidity and intra-MC with different polyelectrolyte doses.

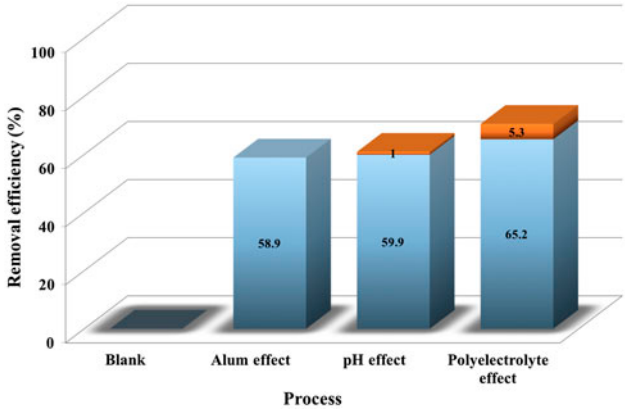

Fig. 4. The effect of pH and polyelectrolyte on *M. aeruginosa* cell removal.

coagulation was improved by 5.3% by combining polyelectrolyte. It shows that a small amount of polymer and pH change could slightly improve intra-MC coagulation by alum.

4. Conclusion

In this work, the optimum operational conditions were determined for the removal of *M. aeruginosa* cell, which is found in drinking water source and is one of the most common types of toxic cyanobacteria through the coagulation process. The effects of chemical dose (coagulant and coagulant aid) and pH on the removal of *M. aeruginosa* cell by coagulation were evaluated with jar tests experiments conducted on synthetic waters. Several different combinations of alum dose, pH, and polyelectrolyte dose were tested to achieve optimum results. *M. aeruginosa* cell concentration was measured and expressed as intra-MC.

According to the results and discussion, the following conclusions have been drawn:

(1) The optimum coagulation conditions for effective removal of intra-MC are a coagulant dose of 60 mg/L, pH value 7.0, and polyelectrolyte dose of 0.2 mg/L.
(2) The best removal efficiency obtained was 65.2% for intra-MC.
(3) In the coagulation process, all intra-MC were not removed with alum. It gave insufficient results in reducing *M. aeruginosa* cells.
(4) Coagulation with higher alum dose may increase aluminum concentration in drinking water.
(5) The pH of the treated water did not present major variation after the coagulation process.
(6) There are interactions between removal of intra-MC and turbidity.
(7) The damage of cyanobacterial cells and the release of intracellular toxin were not reported in this study. This study provided only analysis of intracellular toxin in the alum coagulation. Hence, it is suggested that further investigations should be conducted in order to observe cyanobacterial cell damage and toxin release in response to alum treatment.

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