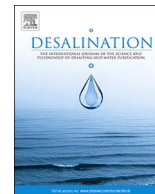




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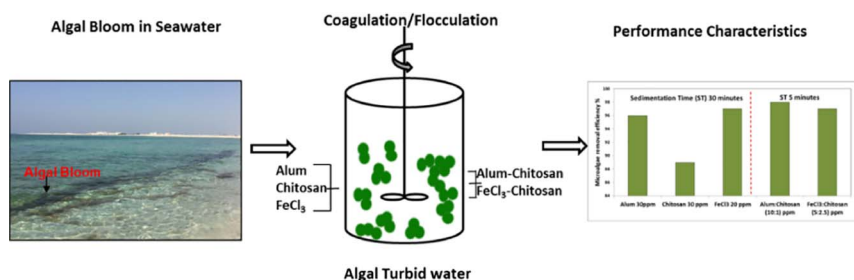
## Desalination

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## Removal of microalgae from seawater using chitosan-alum/ferric chloride dual coagulations

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## GRAPHICAL ABSTRACT



## ARTICLE INFO

## Keywords:

Harmful algal bloom (HAB)  
Sustainable seawater feed  
Microalgae removal  
Dual coagulation  
Bioflocculant  
Coagulation-flocculation-sedimentation process

## ABSTRACT

During algal bloom, it's a challenge to provide good quality feed water, and ensure sustainable RO plant operations without an adequate pre-treatment of seawater. In this paper, the effectiveness of the coagulation process with the individual and dual coagulants, using alum, FeCl<sub>3</sub> and chitosan, were explored aiming to remove microalgae from seawater. The coagulation-flocculation-sedimentation (C-F-S) experiments were conducted by optimizing multiple process strategies to reduce the amounts of coagulants and also to shorten the sedimentation process time. The coagulation-flocculation-dissolved air flotation (C-F-D) experiments were performed to generate the process data in order to evaluate the dual coagulation process performance of the C-F-S system. C-F-S experiments using FeCl<sub>3</sub> coagulant gave better process performance (20 ppm FeCl<sub>3</sub> dose, 8.2 pH, 30 min sedimentation time and 98% microalgae removal efficiency) when compared to alum and chitosan based individual coagulations. The process time of the coagulation process was significantly reduced by the addition of chitosan as a flocculent aid. For dual coagulation using alum (10 ppm) as coagulant and chitosan (1 ppm) as flocculent aid improved microalgae removal efficiency to 98% at a reduced process time of 5 min, making C-F-S process as attractive as C-F-D process.

## 1. Introduction

More than half of the world's population lives in water stress areas, and the numbers are expected to increase to two-thirds by 2025. In many parts of the world, shortage of fresh water is a looming crisis due to climate change and the increase of the global population [1].

Seawater desalination is one of the feasible solutions in addressing the water crisis [2–6]. Reverse osmosis (RO) is a widely used technology for desalinating seawater [7]. However, sustainable operations of sea water reverse osmosis (SWRO) plants depend on the quality of the feed water. Seasonal microalgae blooms are one of the operational challenges faced by SWRO operators, where blooms can hamper the performance of the

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plant and potable water quality [8–13]. Apart from inducing particulate fouling on the membrane which results in sporadic plant disruptions [14–17], microalgae has a tendency to release potent toxins into the water when its cells are ruptured due to trans-membrane pressure. These toxins pose severe health problems in humans [18–21], resulting in dermatologic, gastrointestinal, respiratory, and neurologic disorders. The focus of this work is to improve performance of pre-treatment processes to produce microalgae free feed water for SWRO plants.

There are several treatment options available to remove microalgae from feed water, namely: (i) disinfection (ii) filtration and (iii) physico-chemical removal process. Although chlorination is effective in disinfecting microalgae, the presence of residual chlorine can significantly decrease the lifespan of membranes. Similar to chlorination, other disinfection methods, such as UV and ozone, can result in cell-lysis, which releases toxins into the feed water [22]. Sand filtration/flotation processes are ineffective in removing microalgae, unless preceded by chemical coagulation-flocculation. The retention efficiency of a sand filter obtained for 145,000 cells/ml algae was reported as 80% during the first few hours of filtration and dropped to 48% after 7 h [23]. For the coagulation process, alum and ferric chloride are among the common coagulants used by the water treatment industry. Ferric salts are preferred in sea water desalination, due to the low solubility of the resulting ferric hydroxide in seawater, over a wide range of pH. On the other hand, the use of alum as a cationic coagulant in seawater is not favoured due to the high solubility of aluminium hydroxide in seawater, which leads to the precipitative scaling of RO membranes [12]. Natural coagulants have also been used in water treatment processes [24, 25]. Chitosan, a cationic polymer prepared from crab/shrimp shells, is the second most abundant biopolymer in the world after cellulose [26]. Chitosan is positively charged, due to the protonization of amino groups in a solution which makes it attractive for a variety of binding applications.

Following coagulation-flocculation, the dissolved air flotation process (C-F-D) is commonly used in the upstream of MF/UF systems to minimize the solid loadings [27–30]. Dissolved air flotation (DAF) is a relatively quick process which is suitable for the removal of low-density algal particles. However, DAF is an energy intensive process (0.05–0.075 kWh/m<sup>3</sup> of treated water) [31], as the generation of air microbubbles requires compressed air requiring as high as 7 bars. Another option to remove microalgae is by the coagulation-flocculation-sedimentation (C-F-S) process. However, it is less preferred, as a long settling time is required to achieve a comparable removal rate with DAF. Sedimentation time of the C-F-S process using some common coagulants for microalgae removal was reported as > 2 h [32]. Optimization of the coagulation strategies can achieve the best microalgae removal rate in the shortest time possible. The sedimentation time of microalgae can be shortened by improving the size and density of the flocs. The purpose of this article is to explore the C-F-S process, and flocculation properties of the dual coagulants, alum-chitosan, and FeCl<sub>3</sub>-chitosan for microalgae removal in seawater. The dual coagulation strategy was followed to minimize the coagulant dosage, in order to optimize the density of flocs, and to minimize the sedimentation time.

## 2. Materials and methods

In Qatar, the algal bloom season starts in the month of October (Fig. 1B) and ends approximately around the month of April, as shown in Fig. 1A. Seawater samples analysed during these months revealed that the microalgae counts were around six times higher at the start of bloom season, when compared to the April data (Table 1). Phytoplankton counts were calculated by measuring the chlorophyll intensity of each cell, using a BD Accuri C6 flow cytometer (Ann Arbor, Michigan, USA). The dual threshold triggers on FL3 (Excitation: 488 nm; Emission: 670 nm) and FL4 (Ex: 640 nm; Em: 675 ± 12.5 nm) were set just above background noise. A 50 µL sample was injected at the

medium fluidic settings (35 µL/min; core size 16 µm) in order to obtain the absolute cell counts. Particle size measurements were taken by using a Jorin VIPA B HiFlo analyser (Leicestershire, UK), which indicated that the microalgae size was between 2 and 5 µm. In addition to microalgae, macroalgal deposits were found on the shore (Fig. 1 Captions 'C1 and C2'). These blooms can cause ecological, and societal impacts, including the disruption of the intake of water for cooling/desalination [12].

The seawater samples were collected from the west coast of Qatar, in bulk, to prepare the microalgae culture solution. NKP salts were added, as per the literature method [33], on a weekly basis to the sea water in order to enhance algae growth. The purpose of using a sea water culture is to reflect heavy bloom conditions, with microalgae cell counts ~1.5 × 10<sup>3</sup> per µL, and to maintain the consistency of the algae model solution during the testing period.

Jar tests were carried out by using a programmable apparatus (Phipps & Bird, USA) at room temperature. The tester was programmed for rapid mixing at 100 RPM for 1 min; slow mixing at 30 RPM for 15 min; followed by sedimentation for 30 min [34]. Residual aluminium concentration was determined using Agilent inductively coupled plasma-mass spectrometer (ICP-MS 7500c), equipped with automatic sampler introduction and with concentric and microflow nebulization. For dual coagulation using alum, two sets of experiments were conducted as shown in Fig. 2. Firstly, an alum and chitosan mixture was used as a primary coagulant, which was added during the rapid mixing and the coagulation stage. The second set of experiments were conducted using alum as the primary coagulant and chitosan as the secondary coagulant which acted as a flocculant aid. The primary coagulant was added during the rapid mixing stage, and the secondary was added during the slow mixing stage.

About 100 mL of samples were taken after the sedimentation stage for analyses. Similar experiments were conducted using FeCl<sub>3</sub> as the primary coagulant, and chitosan, as a flocculant aid.

DAF jar tests were performed using the batch jar tester Platypus DAF system, with a 2 L capacity DAF saturator. For DAF particle separation experiments, coagulation and flocculation processes were followed by a 10 min period of flotation, using a 15–30% recycling ratio at a saturation pressure of 675 kPa [28]. The operational conditions of coagulation/flocculation are similar to the sedimentation studies. The DAF-treated samples were collected via sampling ports for characterization studies (microbial counts, turbidity). The C-F-D process was performed using alum and FeCl<sub>3</sub> as coagulants, mainly to generate the process data in order to evaluate the dual coagulation process performance of the C-F-S system.

## 3. Results and discussion

### 3.1. Coagulation studies with alum, chitosan and FeCl<sub>3</sub>

The jar test experiments were conducted with alum, chitosan and ferric chloride coagulants to determine the optimal coagulant dose, based on settled water turbidity (Fig. 3A). The clarified water turbidity, decreased with coagulant additions to the levels (0.7, 2.7 and 0.5 NTU for alum, chitosan and FeCl<sub>3</sub> respectively), after which turbidity increased. The optimum dose of alum, chitosan, and FeCl<sub>3</sub> for maximum turbidity removal was found to be 30, 30 and 20 mg/L respectively. The corresponding micro algae removal efficiency of alum, chitosan and FeCl<sub>3</sub> were found to be 96.3, 87.3 and 98.6% respectively (Table 2).

An overdose of alum (50 ppm) caused charge reversal which led to decrease in microalgae removal efficiency (Fig. 4 A) and increase in turbidity values. An overdose of chitosan led to an increase in turbidity values, while the microalgae removal efficiency remained constant 87% (Fig. 4B). Furthermore, the higher ferric chloride dose increased the turbidity values; while higher microalgae removal efficiency was observed (Fig. 4E). Results show the importance of both microalgae cells counts, and turbidity measurements, to determine the extent of the

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