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Assessing the application and downstream effects of pulsed mode ultrasound as a pre-treatment for alum coagulation

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ABSTRACT

The application of pulsed mode ultrasound (PMU) as a pre-treatment for alum coagulation was investigated at various alum dosages and pH levels. The effects of the treatments on turbidity and dissolved organic carbon (DOC) removal and residual Al were evaluated. Response surface methodology (RSM) was utilized to optimize the operating conditions of the applied treatments. The results showed that PMU pre-treatment increased turbidity and DOC removal percentages from maximum of 96.6% and 43% to 98.8% and 52%, respectively. It also helped decrease the minimum residual Al from 0.100 to 0.094 ppm. The multiple response optimization was carried out using the desirability function. A desirability value of >0.97 estimated respective turbidity removal, DOC removal and Al residual of 89.24%, 45.66% and ~0.1 ppm for coagulation (control) and 90.61%, >55% and ~0 for coagulation preceded by PMU. These figures were validated via confirmatory experiments. PMU pre-treatment increased total coliform removal from 80% to >98% and decreased trihalomethane formation potential (THMFP) from 250 to 200 ppb CH_3Cl. Additionally, PMU application prior to coagulation improved the settleability of sludge due to the degassing effects. The results of this study confirms that PMU pre-treatment can significantly improve coagulation performance.

1. Introduction

Coagulation is an important process in surface water treatment. It is normally applied for removing turbidity, color, microbes and DOC [1]. Recently, much attention is given for improving coagulation performance particularly for DOC removal owing to the involvement of DOC in causing technical and health problems such as filtration fouling and formation of disinfection by-products (DBPs) [2]. Increasing DOC removal with coagulation requires in-depth understanding of coagulation mechanisms. There are four main mechanisms through which coagulants can scavenge DOC from water: charge neutralisation, adsorption, enmeshment and complexation of DOC with coagulants [3]. In reality, these mechanisms occur concurrently, however, there is always a dominant mechanism depending on the properties of the water and the quantity of the applied coagulant [4]. Based on this principle, the term enhanced coagulation came into existence where the amount of coagulants and pH are adjusted to achieve maximum DOC removal. As mentioned earlier the role of coagulation is not confined to DOC removal only, other forms of contaminations are targeted too (e.g., turbidity). Unfortunately, the optimum conditions for DOC removal do not always align with the removal of other forms of contaminants, making it harder to achieve the desired highest level of DOC removal [3]. Therefore, the application of pre-treatment techniques prior to coagulation is essential to maximize DOC removal.

Oxidation pre-treatment techniques are generally utilized for partial removal of DOC prior to coagulation [5]. Chlorine or chlorine derivatives were traditionally used for pre-oxidation [6], but the risk of DBPs formation led to their replacement with other oxidants such as ozone. However, ozone has also been found to form toxic materials such as aldehydes [7]. Recently, the application of advanced oxidation techniques (AOP) such UV light has emerged as a safe pre-oxidation method for removing DOC prior to coagulation. As the water prior to coagulation can be turbid, the penetration of UV light through it would be weak, resulting in low DOC removal levels. Therefore, the application of another AOP namely ultrasound for improving coagulation performance was proposed in this study.

Applying ultrasound for such a purpose has not been investigated thoroughly in previous works. A very limited number of publications touched on this topic such as the work conducted by

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Ziylan and Ince [8], and these research efforts only went as far as measuring DOC removal when combining ultrasound with coagulation. In this work, the application of ultrasound as a suitable pre-treatment for the most commonly used coagulant (alum) was carefully evaluated. Pulsed mode ultrasound (PMU) was selected for this study due to the high efficiency of this mode in removing water contaminants when compared to standard continuous mode [9,10]. Two treatment types: coagulation alone (control) and coagulation with PMU pre-treatment were applied at various alum dosage and pH levels. In addition to DOC removal, other important treatment criteria such as turbidity removal and residual dissolved Al were measured to assess the overall improvement that ultrasound pre-treatment can introduce to coagulation performance. The dissolved form of Al is of concern to water treatment practices due to its toxicity, involvement in neurological diseases and removal difficulty as compared to other Al species [11,12]. Most countries around the world have set a maximum limit for dissolved Al in their finished drinking water. For example, the maximum limit set for dissolved Al in Australia is 0.2 mg/L [13].

The optimization of the experimental factors applied in the control and PMU with coagulation treatments was performed using centre composite design (CCD) of RSM in Design-Expert software. RSM is an effective optimization tool that facilitates identifying the effective levels of experimental factors with minimal number of experiments [14]. Multiple response optimization was conducted using the desirability function where the optimization criteria were set as both maximum turbidity and DOC removal along with minimum residual dissolved Al concentration. The desirability function of the Design-Expert software has the capacity for simultaneous determination of the optimum levels of experimental factors for several responses [15]. The levels of total coliform and THMFP in the treated water with optimum control and coagulation preceded by PMU treatments were measured and compared to gauge the downstream effects of PMU as a pre-treatment technique.

2. Materials and methods

2.1. Water sample

Narda lagoon, situated in the South-East Queensland, Australia was chosen as the sampling site for this study. The samples were collected using 5 L plastic containers. The water samples were sieved through 0.5 mm to simulate the screening required for natural water samples in water treatment practices [16,17]. Detailed information regarding the levels and nature of contaminants in Narda water is shown in Table 1. Table 1 shows that Narda water can be categorized as alkaline hydrophobic water with high microbial load.

Table 1
Water sample properties.

Properties	Levels
pH	7.6
Alkalinity (mg $CaCO_3$/L)	254
Turbidity (NTU)	20
Conductivity (mS/cm), 25 °C	0.26
Total coliform (CFU/100 mL)	250
DOC (mg/L)	9.8
Hydrophobic fraction (%)	78
Hydrophilic fraction (%)	22
$SUVA_{254}$ (L mg^{-1} cm^{-1})	0.036
$SUVA_{280}$ (L mg^{-1} cm^{-1})	0.025

2.2. Pulsed ultrasound treatments

Ultrasound was used in this study as a pre-treatment for the coagulation process. Ultrasonic treatments were carried out using a digital Branson sonifier model 450 with operating low frequency of 20 kHz and a maximum electrical power of 400 W (Branson, USA). The sonifier is equipped with a titanium horn (∅ = 19 mm). The treatments were conducted in batches using 500 mL Pyrex beaker. The temperature of water samples was maintained at approximately 20 °C using a water bath. Power characterization of the ultrasonic system was studied and discussed in our previous work [18]. The water samples were treated by applying pulse ratio (*On:Off*) of 0.2:0.1 s at a calorimetric power of 48 W for an effective treatment time of 5 min. The aforementioned treatment parameters were selected in this study due to their efficient removal of contaminants such as microbes [10].

2.3. Coagulation/flocculation treatments

Coagulation/flocculation treatments were performed using VLEP-Scientifica jar tester (Model JLT 6). The mixing time and speed of coagulation and flocculation processes were set through an electronic register. Six 1 L Pyrex beakers were used for carrying water samples. Alum coagulant prepared from $Al_2(SO_4)_3$ salt and used for water treatment. The important factors for coagulation/flocculation processes are coagulant dosage, pH and the operating parameters of the processes viz. mixing speeds and times. The effective range of coagulant dosage and pH for the tested water were determined through preliminary experiments. The pH of water samples was controlled using 0.1 N HCl and 0.05 N NaOH solutions. The operating parameters for the jar test were selected based on the reported conditions in the literature for the use of alum in surface water coagulation as summarized in Table 2. Water samples were coagulated at 150 rpm for 1 min followed by flocculation at 30 rpm for 15 min and finally were left for 30 min to settle. After settlement, samples of the supernatant were withdrawn at 2 cm depth from the water surface and used for measuring the targeted characteristics such as turbidity, DOC and residual dissolved Al. The downstream effects of ultrasound pre-treatment were evaluated by measuring total coliform and THMFP of the supernatant with and without ultrasound pre-treatment. Prior to total coliform and THMFP measurements, the supernatant samples were filtered through 11 μm filter paper (Whatman, grade 1) to simulate a sand filtration process that is commonly used in water treatment practices [19,20].

2.4. Analytical methods

DOC of water samples was measured by applying the standard high-temperature combustion method as described in [21]. Total carbon analyzer model TOC-V_{CSH} supplied with an auto-sampler (ASI-V) (SHIMADZU, Australia) was utilized for DOC measurements. Three injections were made for each sample resulting in coefficient of variance of less than 0.02.

Residual dissolved Al in the treated samples was measured applying the standard direct nitrous oxide–acetylene flame

Table 2
Ranges of jar test operating parameters reported in relevant literature.

Operating parameters	Range	Refs.
Coagulation speed (rpm)	100–300	[21–27]
Duration of coagulation (min)	1–1.5	[21–27]
Flocculation speed (rpm)	20–40	[21–27]
Duration of flocculation (min)	15	[21,22,24–26]
Settling time (min)	30	[21–23,25,26]