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Effect of bioflocculants on the coagulation activity of alum for removal of trihalomethane precursors from low turbid water

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ABSTRACT

Reactivity of chlorine towards hydrophobic groups present in natural organic matter (NOM) provokes the formation of carcinogenic disinfection byproducts such as trihalomethanes in chlorinated water. The present study aimed to investigate the variations in coagulant activity of alum using two different bioflocculants (coagulant aid) namely, *Moringa oleifera* and *Cyamopsis tetragonoloba* for the removal of hydrophobic fractions of NOM and subsequent chlorine consumption by treated water. Effect of dual coagulants on trihalomethane surrogate parameters such as total organic carbon, dissolved organic carbon, UV absorbing materials and prominent hydrophobic species such as phenolic groups along with aromatic chromophores, polyhydroxy aromatic moiety have also been studied. The concept of differential spectroscopy and absorbance slope index has been employed to understand the combined effects of alum-bioflocculants on the reactivity of NOM with chlorine. Our result shows that the combination of alum and *C. tetragonoloba* is more efficient for reducing trihalomethane surrogates from chlorinated water as compared to *M. oleifera*. *C. tetragonoloba* elicited synchronized effects of sweep coagulation and particle bridging-adsorption which eventually facilitated efficient removal of hydrophobic fractions of NOM. The variation in the mechanistic approach of bioflocculants was due to the presence of cationic charge on *M. oleifera* and adhesive property of *C. tetragonoloba*.

Introduction

The mitigation of natural organic matter (NOM) from surface water has been a major concern pertaining to their concomitant toxicity on human health. During the past 20 years, a notable increase in the amount of NOM has been reported across the globe (Evans et al., 2005; Worrall et al., 2009). The preponderance of NOM due to putrefied terrestrial vegetation and aquatic biota, agricultural runoff, irregular climatic pattern and disposal of anthropogenic wastes have eventually increased the organic load in raw water sources (Fabris et al., 2008; Golfinopoulos, 2013). Excessive NOM deteriorates color, odor, and taste of drinking water, but the intense reactivity of aromatic fractions of NOM towards chlorine has led to the formation of trihalomethanes (THMs) in chlorinated water (Liu et al., 2010). Though, chlorination is the most efficient preventive measure which has marked significant control over dominance of microbial contaminants in drinking water, but the formation of carcinogenic

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Abbreviations: ASI, Absorbance Slope Index; DBPs, Disinfection byproducts; NOM, Natural Organic Matter; DOC, Dissolved Organic Carbon; TOC, Total Organic Carbon; PHA, Polyhydroxy Aromatic; SUVA, Specific UV Absorbance; *M. oleifera*, *Moringa oleifera*; *A–M. oleifera*, Alum–*Moringa oleifera*; *C. tetragonoloba*, *Cyamopsis tetragonoloba*; *A–C. tetragonoloba*, Alum–*Cyamposis tetragonoloba*; MIC, Minimum Inhibitory Concentration.

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disinfection byproducts (DBPs) has emerged as an unexpected consequence in chlorinated water. The detrimental effects of THMs in chlorinated water have raised health concern due to their associated carcinogenic and non-carcinogenic risks (Priya and Mishra, 2017; Mishra et al., 2014, 2016; Zeng et al., 2016).

NOM is often considered as weak anionic polymer due to the presence of charged hydrophobic in the structure (Sweitlik et al., 2004). The structural matrix of NOM includes hydrophobic and hydrophilic components, but hydrophobic groups such as humic acids, fulvic acid and humin have contributed to one-half of dissolved organic carbon (DOC) in natural water (Thurman, 1985). Primarily, hydrophobic fractions of NOM are enriched with conjugated double bonds, aromatic carbon and phenolic structures while the aliphatic groups include aliphatic carbon and nitrogenous compounds, such as carbohydrates, sugars and amino acids (Sweitlik et al., 2004).

The hydrophobic fractions of NOM contain the majority of charged functional groups, thus coagulation could be considered as the amenable approach to reduce the hydrophobicity in NOM enriched raw water.

Coagulation is defined as the process of destabilization of particulate, colloidal and suspended particles dispersed in water using coagulating agents such as inorganic salts (alum, ferric sulfate, ferric chloride, zirconium oxychloride, titanium tetrachloride), synthetic organic polymers (polyaluminium chloride, polyethylene imine) and natural coagulants *(Moringa oleifera, Strychnos potatorum, Aseculus hyppocastanum)* (Jarvis et al., 2012; Priya et al., 2017; Sciban et al., 2009; Muthuraman et al., 2014; Abebe et al., 2016; Song et al., 2016).

The process of particle agglomeration and subsequent sedimentation is based on four basic mechanisms, namely, adsorption and charge neutralization, sweep flocculation, adsorption and interparticle bridging and double layer compression (Miller et al., 2008).

During the process of "adsorption and charge neutralization", positively charged coagulant interact with negatively charged colloidal particles, due to which negatively charged sites get shielded which eventually lead to precipitation and contaminates are adsorbed over the precipitates.

In the case of sweep coagulation, organic and suspended contaminates and natural organic matter get enmeshed inside the porous precipitate of metallic hydroxide formed (Lee et al., 2000; Miller et al., 2008). However, coagulant tends to forms a polymeric chain upon which dispersed particles get adhered during the process of particle bridging (Miller et al., 2008). Thus, characteristics of flocs and treatment efficiency depend on operational condition and mechanistic approach of the coagulation process to a large extent.

Currently, aluminum salt (alum) is the most preferred coagulant due to the efficiency and cost effectiveness but traces of aluminum residuals in treated water have been potent agent for Alzheimer's diseases (Arezoo, 2002). The issue of sludge disposal is another environmental threat during wide application of aluminum salt as primary coagulant. However, researchers have validated that aluminum salt is less effective in removing hydrophobic fractions of NOM as compared to ferric salts and zirconium oxychloride (Jarvis et al., 2012; Priya et al., 2017). The complex structure of synthetic organic polymers has raised concern due to their incomplete biodegradation and monomers of such coagulants like acrylamide are known mutagenic and neurotoxic agents (Dearfield et al., 1988; Kwon et al., 1996).

The use of natural coagulant as coagulant is an economical and effective remediation approach for the treatment of highly turbid water, but the organic matrix of plant tissue microorganism and animal extracts such as glycoprotein, polysaccharides, nucleic acid and protein tends to increase the organic content in water (Verma et al., 2012). Though, the structural composition of natural coagulants assures their non- toxicity and biodegradability properties, but the low flocculating activity with respect to turbidity removal from surface water has also been reported in various studies (Sciban et al., 2009; Muthuraman and Sasikala, 2014). The insignificant flocculant activity effects of natural coagulants in low turbid water might be due to restricted repulsive force between natural coagulant dispersed colloidal particles (Huang et al., 2014). Therefore, low flocculating properties, insignificant yields and high production costs have constraint the field application of natural coagulants in water treatment plants.

Various researchers have explored the scope of "Enhanced coagulation" using dual coagulants as an attempt to overcome the limitation of inorganic and natural coagulants. The enhanced coagulation is the effective concept for improving the efficiency of primary coagulant using a natural coagulant as bioflocculants for water treatment under optimized operational condition. The combined effects of charge neutralization, sweep coagulation and flocs bridging drives the process of coagulation using metal coagulant and bioflocculants, which have shown pronounced effects on coagulant activity (Bo et al., 2012). Researchers have studied the combined effects of bioflocculants MBFGA1 and polyaluminium chloride and observed that dosages of metal coagulant reduced, and the coagulation behavior increased in treated water (Yang et al., 2009). The robust flocs forming efficiency of aluminum sulfate and bioflocculants in kaolin-humic acid solution had also been reported (Bo et al., 2011).

In this study, the scope of enhanced coagulation has been explored to investigate the feasibility of alum-bioflocculants (*Cyamopsis tetragonoloba* (*C. tetragonoloba*) and *M. oleifera* (*M. oleifera*)) mediated coagulation process for reduction of total organic carbon (TOC), DOC, specific UV absorbance (SUVA), absorbance slope index (ASI) and differential spectroscopy (ΔA272 nm) from NOM enriched synthetic water. The performances of dual coagulants have also been evaluated based on their ability to degrade hydrophobic species such as phenol groups from NOM and subsequent reduction in chlorine consumption. The variation in the mechanistic approach of bioflocculants has been elucidated based on their morphology and chemical characterization.

1. Material and methods

1.1. Reagents

Aluminum sulfate octadecahyderate ($Al_2(SO_4)_3{\cdot}18H_2O$), (HCl) and NaOH was procured from Merck, India. Humic acid and kaolin were purchased from Loba Chemie, Mumbai, India. Seeds of *M. oleifera*, *C. tetragonoloba* extract (powder) and nutritional agar were supplied by Sun Seeds Pvt. Ltd., Akshar Chemicals and Hi

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