



Toward a better understanding of coagulation for dissolved organic nitrogen using polymeric zinc-iron-phosphate coagulant



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ABSTRACT

The increase of agricultural related activities and the lack of effective waste control has led to an increase of organic nitrogen in water. The development of coagulants to effectively remove dissolved organic nitrogen (DON) is a high priority in the water treatment industry. We developed a polymeric zinc-iron-phosphate (Zn-Fe-P) coagulant and investigated its coagulation effect on DON removal. Optimum coagulant for coagulation for DON and TDN removals was characterized by the dense convex-concave packing structure differing from other zinc-based coagulant, polycrystalline structure and high content colloidal species, which could account up to 87% of the total colloidal species. Coagulation experiments showed the DON removal rate to vary greatly depending on principal components and their interaction with metals, phosphate and hydroxyl. DON removal efficiency increased with the increase of colloidal species. The coagulation was also dependent on coagulant dosage and water quality parameters: Coagulation efficiency increased with coagulant dosage in the investigated range of 1–16 mg/l, and a pH of 6 was found to be superior for the coagulation. DON removal efficiency was also higher than and linearly correlated with total dissolved nitrogen (TDN) removal, which implies that an effective coagulation for TDN is also effective for DON. The findings in this study indicate that coagulation of DON is largely influenced by coagulant composition and species. We also found the removal of DON by our newly developed polymeric Zn-Fe-P coagulant to be effective.

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1. Introduction

The coagulation-flocculation process is an essential part of water treatment, especially for drinking water treatment. As a viable option for the treatment of surface water, it is able to remove suspended solids, colloidal particles, natural organic matter (NOM) (Amuda and Alade 2006, Matilainen et al. 2010, Keeley et al. 2016). During this process, it is common to observe higher removal efficiencies for hydrophobic fractions of NOM and high molecular mass compounds than for hydrophilic fractions and low molecular mass compounds (Matilainen et al. 2010, Zhu et al. 2014). The hydrophilic fraction contains higher aliphatic content and nitrogen content

carboxylic acids, carbohydrates, and proteins (Matilainen et al. 2010, Li et al. 2014). Therefore, it is expected that nitrogenous compounds will be difficult to remove by the conventional coagulation-flocculation process. Although the dissolved organic nitrogen (DON) only comprises a small portion of dissolved organic matter (DOM) in surface water (Gur-Reznik et al., 2008) (about 0.5–10% by weight), it could result in harmful nitrogenous disinfection byproducts (N-DBPs) during chlorination and chloramination processes (Lee et al., 2007). The N-DBPs present higher genotoxic, cytotoxic, and carcinogenic effects than the conventional carbonaceous DBPs such as trihalomethanes and haloacetic acids (Shah and Mitch, 2012). In addition, DON is one of the important influential factors on membrane fouling (Zhang et al., 2006). In order to keep water sources reliable and clean, research on technologies that can remediate contaminated water and prevent further DON pollution to the aquatic environment has received

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considerable attention (Gu et al., 2014; Czerwionka and Makinia, 2014; Zeng et al., 2014).

The mechanisms for NOM removal by coagulation-flocculation are usually assigned to charge neutralization, adsorption bridging, and sweep flocculation (Li et al., 2006). In the presence of hydrolysable metal ions such as trivalent aluminum (Al (III)), trivalent ferric iron (Fe (III)) or polyelectrolyte with opposite charge to the colloid surface, the surface charge of colloids could be neutralized by adsorbing metal hydrolysis species onto their surface. The flocculation of negative colloidal particles with cationic coagulant could result in large flocs, followed by sedimentation. With the common aluminum/ferric (Al/Fe) coagulant, coagulation-flocculation has been considered to be an inefficient process for DON removal (Lee and Westerhoff, 2006; Gu et al., 2010). Although the inorganic-organic coagulant has been reported to improve the DON removal rate (Lee and Westerhoff, 2006), the efficiency is still not high enough for practical application. Therefore, combined application of coagulation-flocculation with other technologies have also been reported (Arnaldos and Pagilla 2010). Although numerous efforts have been devoted to the removal of carbon and phosphorus compounds, a paucity of research has been devoted to the removal of nitrogenous compounds.

Coagulation-flocculation behavior can be affected by many parameters including coagulant type and dosage as well as ambient conditions (Chakraborti et al. 2003). Of these, the coagulant type is the major factor influencing coagulation-flocculation efficiency. The development of coagulant types focuses on preparing multiple metal complexes, especially during the complexing of Al, Fe and silicon (Si) (Gao et al. 2003, Zhu et al. 2011). Recent research revealed that the complexing of zinc ion (Zn (II)) with other metal ions had a superior performance than some common coagulants (e.g., polymeric ferric sulfate and polymeric aluminum sulfate). These coagulants' speciation and morphology have been known to affect coagulation behavior significantly. Three typical types of zinc composite coagulants have been developed including poly-ferric-zinc-sulfate (PFZS) (Wei et al. 2015), poly-zinc-silicate-sulfate (PZSS) (Zeng and Park 2009) and poly-aluminum-zinc-ferric (PAZF) (Ying et al. 2012). Although the presence of Zn (II) is more efficient for removing ammonia nitrogen ($\text{NH}_4\text{-N}$) by producing hydroxides of zinc (Ying et al., 2012) or other metallic salts (e.g., magnesium ammonium phosphate (Fattah et al., 2013), further research on their coagulation for DOM is needed.

To better understand coagulation for DON by polymeric Zn-Fe coagulant, a hydrolyzed zinc-iron-phosphate (Zn-Fe-P) coagulant (denoted as PPZFS) in this study was developed. PPZFS was synthesized through hydrolysis and polymerization of Zn (II), Fe (III) and phosphate (PO_4^{3-}) ions. The difference from previous preparation is that the PO_4^{3-} introduced in PPZFS has a relatively weaker affinity for Fe (III) than hydroxyl (OH^-) but stronger than other anions (e.g., F^- , SO_4^{2-} and Cl^-), which can compete with OH^- for partial binding sites of Fe (III) hydrolysis products, adjusting its hydrolytic/polymeric species thus improving coagulation behavior to yield a more efficient removal of contaminants (Tang 2006). The coagulant was characterized via Fourier translation infrared spectroscopy (FT-IR), scanning electron microscope (SEM) and X-ray diffraction (XRD). The species distribution in PPZFS was measured by a timed complexation spectroscopy known as the Ferron method (Hu et al., 2001). Coagulation effects on removal of DON from polluted source water were systematically examined. Analysis was performed using a scientific software (Version 8.0, OriginLab).

2. Materials and methods

2.1. Materials

All reagents used in this study were primarily of analytical grade, including ferrous sulfate ($\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$), sodium acetate, zinc sulfate (ZnSO_4), potassium dihydrogen phosphate (KH_2PO_4), sodium hydroxide (NaOH), sodium chlorate, sodium carbonate (Na_2CO_3), concentrated sulfuric acid (H_2SO_4), and Ferron reagent (8-hydroxy-7-iodoquinoline-5-sulfonic acid). Iron and zinc powder were guarantee reagents. All aqueous solutions and standard solutions were prepared with deionized water. The glassware and other labware were acid-washed, rinsed thoroughly with water, and dried prior to use.

2.2. Coagulant preparation

PPZFS was prepared using sodium chlorate as the oxidant. First, 54.3 g of $\text{FeSO}_4 \cdot 7\text{H}_2\text{O}$ was acidified by concentrated sulfuric acid, followed by slow stirring in a beaker until a homogeneous liquid mixture was obtained. Second, a certain amount of sodium chlorate solution (3M) was added into the reaction vessel. Then, ZnSO_4 powder with a pre-determined Zn/Fe molar ratio ($n\text{Zn}/n\text{Fe} = 0.1\text{--}1$) was added into the reaction vessel, resulting in the formation of a new liquid mixture containing zinc and iron. The new liquid mixture was stirred rapidly for 1 min followed by 10 min of slow stirring in a thermostatic water bath at a temperature ranging from 30 to 80 °C, to improve the hydrolysis and polymerization reactions. KH_2PO_4 was slowly added into the reaction system as a stabilizer with a pre-determined k value ($k = \text{P/Fe}$ molar ratio as $n\text{P}/n\text{Fe}$ in the range of 0.05–0.55). After the addition of KH_2PO_4 for 10 min, Na_2CO_3 powder was added to the above solution to obtain the desired r value ($r = \text{OH/Fe}$ molar ratio as $n\text{OH}/n\text{Fe}$ in the range of 0.03–0.39). The PPZFS was aged for at least 24 h at room temperature prior to usage.

To obtain an optimized PPZFS, the preparation was modified via the response surface methodology (RSM) by finding the relationship between $n\text{Zn}/n\text{Fe}$, $n\text{P}/n\text{Fe}$ and $n\text{OH}/n\text{Fe}$, and the DON removal efficiency at three levels of each factor using Box-Behnken experimental design. The three levels corresponding to each factor were 0.1, 0.13 and 0.17 for $n\text{Zn}/n\text{Fe}$, 0.35, 0.45 and 0.55 for $n\text{P}/n\text{Fe}$, and 0.06, 0.13 and 0.26 for $n\text{OH}/n\text{Fe}$. An optimized coagulant (denoted as PPZFS_m) was herein obtained.

For comparison, other coagulants including polymeric ferric sulfate (PAS), phosphate-aluminum sulfate (PPAS), polymeric silicon ferric sulfate (PSiFS), polymeric phosphate-aluminum chloride (PPAC), organic-inorganic composite coagulant (PPZFS-polyacrylamide, PPZFS-PAM) and polymeric aluminum ferric sulfate (PAFS) were prepared in our lab and their coagulation effects on DON or TDN removal were also examined.

2.3. Coagulation test

A water sample was collected from Xiang Jiang River located in Xiangtan City, China. For a comparison, those samples from other sites such as streams and lakes around the Xiang Jiang River were also collected. A coagulation-flocculation experiment was performed using a program-controlled jar test apparatus (ZR4-6, Zhongrun Water Industry Technology Development Co. Ltd., China) at room temperature. One liter of collected water sample was transferred into a beaker and the initial pH of the sample was adjusted to the set value using 0.5 mol/l HCl and 0.5 mol/l NaOH. The sample was rapidly mixed at the set agitation speed (rpm) for 2 min, followed by a slow mixing phase at 70 rpm for 15 min, and then a 30 min settling time. The supernatant sample was extracted

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