



A comparative study of ozonation, iron coated zeolite catalyzed ozonation and granular activated carbon catalyzed ozonation of humic acid



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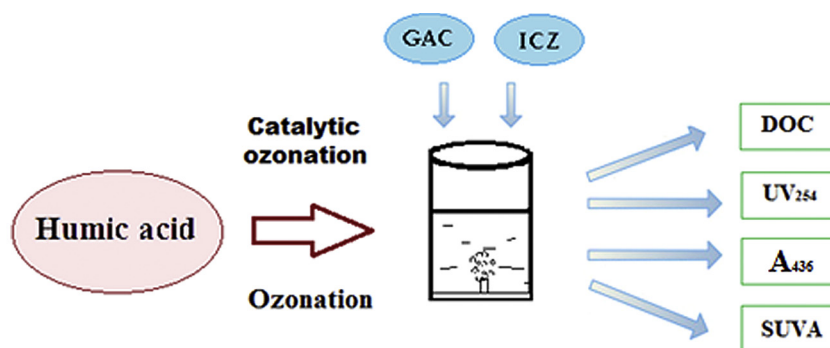
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HIGHLIGHTS

- The performance of iron coated zeolite and granular active carbon for catalytic ozonation of humic acid was evaluated.
- The presence of ICZ and GAC significantly enhanced the degradation of HA.
- Ozonation and catalytic ozonation at pH 11 increased DOC and UV₂₅₄ removals compared to ozonation at pH 6.5.
- Catalytic ozonation with ICZ at pH 11 showed the best catalytic activity for HA removal.
- DOC and UV₂₅₄ removals in ozonation of <100 kDa fraction of HA were better than that of <50 kDa fraction of HA.

GRAPHICAL ABSTRACT



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ABSTRACT

This study compares ozonation (O₃), iron coated zeolite catalyzed ozonation (ICZ–O₃) and granular activated carbon catalyzed ozonation (GAC–O₃) for removal of humic acid from an aqueous solution. The results were evaluated by the removal of DOC that specifies organic matter, UV₂₅₄ absorbance, SUVA (Specific Ultraviolet Absorbance at 254 nm) and absorbance at 436 nm. When ozonation was used alone, DOC removal was 21.4% at an ozone concentration of 10 mg/L, pH 6.50 and oxidation time of 60 min. The results showed that the use of ICZ or GAC as a catalyst increased the decomposition of humic acid compared to ozonation alone. DOC removal efficiencies were 62% and 48.1% at pH 6.5, at a catalyst loading of 0.75 g/L, and oxidation time of 60 min for ICZ and GAC, respectively. The oxidation experiments were also carried out using <100 kDa and <50 kDa molecular size fractions of humic acid in the presence of ICZ or GAC. Catalytic ozonation also yielded better DOC and UV₂₅₄ reduction in both <50 kDa and <100 kDa fractions of HA compared to ozonation.

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1. Introduction

Chlorination is the most widely used disinfection practice during water treatment, because it is effective against most pathogens and provides residual disinfecting capacity (Li et al., 2016). However, chlorine can react with natural organic matter (NOM) to generate disinfection byproducts (DBPs). Some of these byproducts are considered genotoxic, mutagenic and/or carcinogenic with obvious adverse effects on human health (Fan et al., 2014).

Trihalomethanes (THMs) and haloacetic acids (HAAs) are the two most abundant groups of DBPs found in chlorinated waters. Many other halogenated DBPs have also been identified in drinking water, including haloacetonitriles, haloketones, and halonitromethanes. Although large in number, the newly identified DBPs are typically present at much lower concentrations than THMs and HAAs (Hua et al., 2015). The US Environmental Protection Agency (USEPA) regulates levels of the four brominated/chlorinated THMs (trichloro-, tribromo-, bromodichloro- and dibromochloromethane, often called THMs), at $80 \mu\text{g L}^{-1}$, and five brominated/chlorinated HAA species (chloro-, bromo-, dichloro-, dibromo- and trichloro-acetic acid, HAAs), at $60 \mu\text{g L}^{-1}$, in drinking water. The THMs are also regulated in EU drinking water at $100 \mu\text{g L}^{-1}$ (as THMs) (Bond et al., 2014).

In recent years, new aromatic halogenated DBPs such as dihalo-4-hydroxybenzaldehydes, dihalo-4-hydroxybenzoic acids, dihalosalicylic acids, and trihalo-phenols have also been identified (Pan and Zhang, 2013). These aromatic halogenated DBPs were found to be decomposed to form commonly known DBPs such as THMs and HAAs during chlorination. Pan and Zhang (2013) reported that source water containing NOM with a higher aromatic content (i.e., a higher specific ultraviolet absorbance) generates higher levels of these new aromatic halogenated DBPs during chlorination, and subsequently transforms higher levels of THMs and HAAs after prolonged chlorination.

The natural organic matter (NOM) composed mainly of humic substances serves as the major precursor to the formation of DBPs. Aquatic NOM is a complex mixture of heterogeneous organic compounds varying in size, structure and functionality from source to source (Hua et al., 2015). It has been proved that the formation of DBPs depends on the concentration of NOM which is the main agent in the reaction. In general, increasing NOM levels in chlorinated water increases the formation of DBPs (Xie, 2016). Conventional processes for water treatment including clarification, coagulation, flocculation, sedimentation, filtration and disinfection only remove 30% of the trihalomethanes precursors (Asgari et al., 2012). Removal of NOM is compulsory prior to disinfection in water treatment plant due to the increasingly stringent limit of disinfection byproducts (Sanly et al., 2008).

Briefly, high HA concentrations and aromatic structures have been associated with high DBPs concentrations and the control and removal of NOM is one of the most important approaches typically applied in water treatment facilities to reduce the formation of DBPs. Therefore, the improvement of processes for the TOC removal is necessary to provide high quality drinking water. In order to remove NOM to the greatest possible extent, increasing attention is nowadays being paid to various oxidation treatments (Jelena Molnar et al., 2012).

Ozone has the proven ability to decrease the concentration of DBPs precursors and a number of microorganisms, and reacts with organic substances to increase their biodegradability. Ozone reacts directly with NOM by electrophilic addition to double bonds, which is a very selective reaction. In addition, indirect reactions take place via hydroxyl radicals, which are one of the most powerful oxidants known, and are generated by ozone decomposition in water. One advanced oxidation process (AOP), based on the generation of

highly reactive hydroxyl radicals, is the so-called heterogeneous catalytic ozonation. These processes increase the production of hydroxyl, superoxide and hydroperoxyl radicals, which have a higher oxidation potential than ozone. Thus the efficiency of the ozonation processes increases (Asgari et al., 2012; Jelena Molnar et al., 2012).

It was reported that activated carbon used together with ozone could provide better removal of color than when the two techniques were applied separately (Nawrocki and Kasprzyk-Hordern, 2010). The effect of the presence of activated carbon in the ozonation process is reported to be mainly the acceleration of the decomposition of dissolved ozone into free hydroxyl radicals (HO^\bullet), which are highly strong oxidant species able to react rapidly with most of the water pollutants in such an advanced oxidation process (AOP) first named as Carbozone (Alvarez et al., 2006).

Natural zeolites are hydrated aluminosilicate minerals of a porous structure with valuable physicochemical properties, such as cation exchange, molecular sieving, catalysis and sorption. In recent years, natural zeolite and its modified forms have also been reported for removal of anions and organics from water systems (Wang and Peng, 2010). However, the catalytic performance of ICZ in ozonation of HA has not been studied.

The aim of this study is to compare ozonation, iron coated zeolite (ICZ) catalyzed ozonation and granular activated carbon (GAC) catalyzed ozonation in the removal of humic acid from an aqueous solution. The catalytic ability of ICZ and GAC in the ozonation of humic acid was evaluated. Furthermore, considering the complexity and polydispersity of the humic macromolecules, they were fractionated into subcomponents according to molecular sizes using ultrafiltration through membranes of 50 and 100 kDa. The effect of ozonation and catalytic ozonation on the molecular size fractions of humic substances was also evaluated.

2. Materials and methods

2.1. Chemicals

Commercially sourced humic acid sodium salt used in all experiments was purchased from Sigma-Aldrich (Sigma-Aldrich Co. Ltd., USA). 1000 mg/L humic acid solutions were prepared by adding 1 g humic acid into distilled deionized water and dissolved using the ultrasonic sonication bath in order to provide homogeneous dissolution. A stock solution of humic acid was stored in amber glass bottles and protected from sunlight. 30 mg/L humic acid was prepared from the stock solution by dilution and filtered through 0.45 μm membrane filter before being used in the experiments. Surface and ground water contains DOC in the range of 2–10 mg/L (Thurman, 2012). So, in this study, HA concentration of 30 mg/L which contains 8.52 mg/L of DOC was used. The characteristics of HA solution of 30 mg/L were given in Table 1.

GAC was obtained from Merck (1.5-mm diameter and 971.7 m^2/g specific surface area). GAC was thoroughly washed with distilled deionized water, dried at 105 °C for 6 h and stored in a desiccator until use in catalytic ozonation experiments.

Table 1
Characteristics of Humic Acid Solution (30 mg/L).

Characteristics	Value
TOC (mg/L)	10.41
DOC (mg/L)	8.52
UV ₂₅₄ cm^{-1}	0.709
SUVA ₂₅₄ L/mg	8.32
A ₄₃₆ cm^{-1}	0.129
pH	6.50

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