



Humic acid removal from micro-polluted source water in the presence of inorganic salts in a gas-phase surface discharge plasma system



Qianrou Zhang^{a,b}, Guangzhou Qu^{a,b}, Tiecheng Wang^{a,b,*}, Chungang Li^c, Hong Qiang^{a,b}, Qihong Sun^b, Dongli Liang^{a,b}, Shibin Hu^{a,b}

^a College of Natural Resources and Environment, Northwest A&F University, Yangling, Shaanxi Province 712100, PR China

^b Key Laboratory of Plant Nutrition and the Agri-environment in Northwest China, Ministry of Agriculture, Yangling, Shaanxi 712100, PR China

^c Ningxia Pengdaxin Energy Environmental Protection Technology Co., Ltd., Yinchuan 750000, PR China

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ABSTRACT

Humic acid (HA), a predominant type of natural organic matter in surface water, had several potential environmental and health risks. A gas phase surface discharge plasma system was employed to eliminate HA from micro-polluted surface water, and the effects of some concomitant inorganic ions on HA removal were evaluated. The experimental results showed that approximately 89.4% of HA could be smoothly eliminated after 40 min of the discharge plasma treatment, and the HA removal process could be fitted well by the first-order kinetic model. The presence of anions such as CO_3^{2-} and Cl^- in the natural water were adverse for the HA removal due to their competition for the active species, whereas the effects of NO_3^- and SO_4^{2-} on the HA removal could be neglected; this deduction was also confirmed via the evolution of fluorescence spectra intensity of $\cdot\text{OH}$ radicals. Although Cu^{2+} could react with H_2O_2 via Fenton-like reaction, its presence was still adverse for the HA removal due to the complexing effect of Cu^{2+} with HA molecules. In addition, the HA removal processes were characterized by UV-Vis spectrum, DOC removal, specific absorbance parameters, and three-dimensional fluorescence spectrum analysis.

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1. Introduction

As natural organic matters, humic substances were extensively present in natural waters, sediments and soils. Humic acid (HA) was the major fraction of the humic substances, which was a poly-electrolytic macromolecule with a flexible molecular weight [1]. HA was the decay and decomposition of plant and animal corpses, which could cause several environmental problems in drinking water [2]. On the one hand, the yellow-to-black colored HA could result in undesirable taste and odor to water environment [3]; on the other hand, it could chelate with some metal ions like Ca^{2+} and Cu^{2+} because a large number of functional groups such as phenolic, carboxyl, carbonyl and hydroxyl groups were in its molecular structure, and their products were one of the etiological factors of Kaschin-Beck disease [4]; even worse, HA was a major precursor of cancerigenic disinfection byproducts in the chlorination process of the drinking water [5]. Thus, it was significant to eliminate the HA from the natural waters.

* Corresponding author at: College of Natural Resources and Environment, Northwest A&F University, Yangling, Shaanxi Province 712100, PR China.

E-mail address: wangtiecheng2008@126.com (T. Wang).

Conventional methods including coagulation [6], adsorption [7], Fenton oxidation [8], TiO_2 photocatalysis [9], and biological treatment [10], membrane filtration [11], UV-photolysis [12], and ozonation [13] have been widely attempted to eliminate HA from the water environment; however, these methods had some disadvantages for HA removal from the micro-polluted source water. For instance, metallic compounds were used in the coagulation process, and some metal ions might enter the treated water and then brought potential risks for the drinking water. Adsorption was just the phase transfer of pollutants from liquid to solid phase, and the pollutants still existed in the adsorbent, and thus the adsorbent needed to be regenerated; in addition, the adsorption capacity of the adsorbent was easily limited by the HA due to its large molecular weight. As advanced oxidation processes, Fenton oxidation and TiO_2 photocatalysis could both efficiently eliminate HA, whereas the $\text{Fe}^{2+}/\text{Fe}^{3+}$ or nano- TiO_2 might bring secondary pollution for the micro-polluted source water. Biological treatment was usually applied for wastewater treatment, and it was not suitable for the micro-polluted source water treatment because some microorganisms were able to release into the water environment. Membrane fouling was the biggest problem for membrane filtration. The potentials of UV-photolysis were generally quite low and thus it was usually combined with some other oxidants such

as H_2O_2 . The reactions between ozone with pollutants were very selective, and it could react with Br^- in natural waters to form Bromate because high ozone concentration provided by traditional ozone generator. Therefore, it was imperial to develop an effective method to eliminate HA from the natural waters.

Recently, dielectric barrier discharge (DBD) plasma has received lots of attention for the removal of organic pollutants [14–19]; in the DBD plasma process, lots of active species such as $\cdot\text{OH}$, $\cdot\text{O}$, H_2O_2 , and O_3 would be generated, allowing the organic pollutants in the wastewater to be decomposed effectively. However, the published literature on the organic pollutants removal by the DBD plasma was mainly focused on the pollutants in the wastewater, and little has been reported on the pollutants removal from the micro-polluted source water. In our previous research, a gas phase surface discharge plasma reactor was developed and its performance for HA removal from the micro-polluted source water was firstly investigated [20]; in this system, reactive species such as O_3 and $\cdot\text{O}$ were first generated in the gas phase, and then they were rapidly injected into liquid phase, and thus short-lived reactive species could be utilized for pollutants removal; more importantly, few risks would be brought during micro-polluted source water treatment by this system because of low concentrations and short lifetimes of these reactive species, and therefore it might be a relatively efficient and safe option for micro-polluted source water treatment, compared with some other methods. For example, some risks would be brought by residual Al^{3+} during coagulation [6]; large amounts of disinfection by-products would be brought by Fenton or ozone treatment [8,13]; nano- TiO_2 might also bring secondary pollution to source water [9]. The safety problems of drinking water in countryside of China were quite serious, especially in hilly regions, and therefore, the gas phase surface discharge plasma was expected as an approach to enhance drinking water quality for the hilly regions.

Inorganic salts such as NaCl , Na_2SO_4 , Na_2CO_3 , and NaNO_3 were always maintained in the micro-polluted source water. The presence of some inorganic salts in the water environment could affect ozone and $\cdot\text{OH}$ reactions [21–23]. Tehrani-Bagha et al. [21] reported that the presence of Na_2CO_3 promoted the decolorization of Reactive Blue 19, whereas NaCl and Na_2SO_4 had negative effects due to their consumption on ozone. Similar phenomena were also found by Muthukumar and Selvakumar, in whose research the presence of NaCl and Na_2SO_4 decreased the decolorization efficiency during the ozonation process [22]. Oppositely, Silva et al. [23] found that the presence of NaCl promoted the self-decomposition of ozone and then enhanced the decolorization efficiency of Acid Red 27. That is, the inorganic salts exhibited different behaviors in the pollutants removal via oxidation process. However, the effects of inorganic salts on HA removal from the micro-polluted source water by the gas phase surface discharge plasma was still unknown.

The aim of this study was to evaluate the HA removal characteristics by gas phase surface discharge plasma in the presence of some inorganic salts. HA removal efficiency and active species ($\cdot\text{OH}$ radical and H_2O_2) generation at different amounts of inorganic salts were investigated. The decomposition of HA was also characterized by UV–Vis, Dissolved organic carbon (DOC), and excitation-emission matrix (EEM) fluorescence analysis.

2. Materials and methods

2.1. Materials

HA was purchased from Aladdin Industrial Corporation, China, which was extracted from peat coal and contains 58.3% of C, 4.2% of H, and 36.1% of O. The average molecular weight of the HA

was 2485 Da. The contents of total acidity, carboxylic and phenolic groups of the HA were 5.92, 2.62, and 3.58 mmol g^{-1} , respectively. NaCO_3 , NaSO_4 , NaNO_3 , NaCl , and CuSO_4 were all analytical-grade and purchased from Guangdong Guanghua Chemical Factory Co. Ltd.

2.2. Surface discharge plasma reactor system

The schematic diagram of the surface discharge plasma reactor system was shown in Fig. 1, which consisted of a power supply, reactor and electrical measurement systems. The power supply was an alternating current power supply with the frequency of 50 Hz. The reactor was a plexiglas cylinder (5 cm inner diameter and 35 cm height). The high voltage electrode was a stainless steel spring with 1 mm wire diameter and attached tightly to a cylindrical quartz tube in its inside wall, and the length of the stainless steel spring was 18 cm. Treated solution was used as the ground electrode. Dry air was bubbled into the treated solution, and the air flow rate was 160 L h^{-1} . The volume of the treated solution was 500 mL, and the HA initial concentration was 30 mg L^{-1} . The discharge voltage was 19.6 kV.

2.3. Methods and analysis

Tektronix TDS2012 digital oscilloscope was employed to monitor the discharge voltage and current signals with a high voltage probe (Tektronix P6015A) and a current probe (Tektronix P6021), and then the discharge power was calculated using the Lissajous method [17],

$$P = \frac{1}{T} \int_0^T UI dt = \frac{C}{T} \int_0^T U \frac{dUc}{dt} dt = fC \oint UdUc = fCS \quad (1)$$

where P was the discharge power, U was the discharge voltage, I was the current, T was the discharge period, f was the discharge frequency, C was the capacitance ($1 \mu\text{F}$), S was the Lissajous figure area and Uc was the voltage over the capacitor. In this study, the discharge power was 21.2 W.

UV–Vis spectrophotometer (UV-1901, Shanghai, China) was used to measure the concentration of HA at a wavelength of 254 nm. Dissolved organic carbon (DOC) was measured by a Total Organic Carbon analyzer (TOC-UCPH, Shimadzu). The

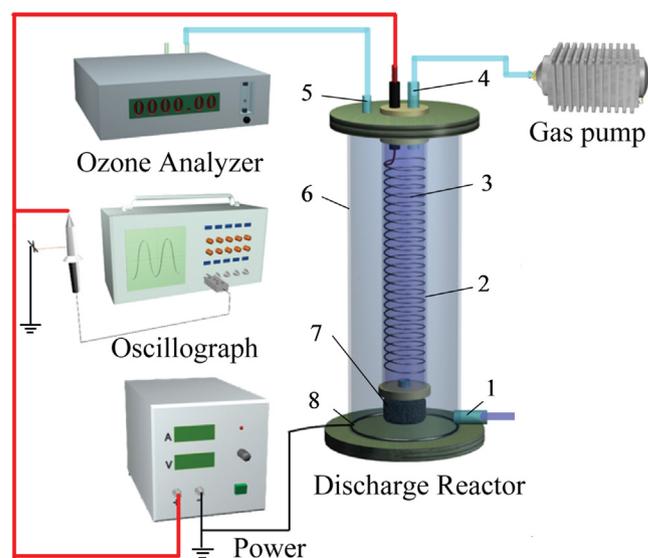


Fig. 1. Schematic diagram of discharge plasma reaction system (1 sampling port; 2 quartz tube; 3 spiral electrode; 4 gas inlet; 5 gas outlet; 6 Plexiglas cylinder; 7 aerator; 8 ground electrode).

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