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Research on the degradation mechanism of pyridine in drinking water by dielectric barrier discharge

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ABSTRACT

Pyridine, an important chemical raw material, is widely used in industry, for example in textiles, leather, printing, dyeing, etc. In this research, a dielectric barrier discharge (DBD) system was developed to remove pyridine, as a representative type of nitrogen heterocyclic compound in drinking water. First, the influence of the active species inhibitors tertiary butanol alcohol (TBA), HCO_3^- , and CO_3^{2-} on the degradation rate of pyridine was investigated to verify the existence of active species produced by the strong ionization discharge in the system. The intermediate and final products generated in the degradation process of pyridine were confirmed and analyzed through a series of analytical techniques, including liquid chromatography–mass spectrometry (LC–MS), high performance liquid chromatography (HPLC), ion chromatography (IC), total organic carbon (TOC) analysis, ultraviolet (UV) spectroscopy, etc. The results showed that the degradation of pyridine was mainly due to the strong oxidizing power of ozone and hydroxyl radical produced by the DBD system. Several intermediate products including 3-hydroxyl pyridine, fumaric acid, 2, 3-dihydroxypyridine, and oxalic acid were detected. Nitrogen was removed from the pyridine molecule to form nitrate. Through analysis of the degradation mechanism of pyridine, the oxidation pathway was deduced. The study provided a theoretical and experimental basis for the application of DBD strong ionization discharge in treatment of nitrogen heterocyclic compounds in drinking water.

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Introduction

Nitrogen heterocyclic compounds are common contaminants found widely in coking wastewater, pharmaceutical wastewater, pesticide wastewater and printing and dyeing wastewater (Zalat and Elsayed, 2013). Discharge of these contaminants into water bodies will cause serious pollution of the aquatic ecological environment. The compounds are classified as bio-refractory compounds and have carcinogenic, teratogenic and mutagenic

characteristics, which are a serious threat to drinking water security (Singh and Chandra, 2005; Li et al., 2001). Pyridine is widely used in industry as a good organic solvent. Meanwhile, pyridine dissolves easily and can exist in natural water stably or infiltrate into groundwater through the soil. Long-term intake of drinking water containing pyridine will cause serious damage to human reproductive and nervous systems (Fetzner, 1998). Pyridine is also a major cause of stench in the air and the occurrence of odor problems in drinking water. Thus, pyridine is

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incorporated in the Surface Water Environmental Quality Standards as part of a drinking water source specific monitoring project in our country, and pyridine compounds are also listed as priority control pollutants by the United States Environmental Protection Agency.

At present, studies on methods for the degradation of pyridine mainly focus on the development of microorganisms, adsorption media, and microbial degradation and catalytic oxidation processes (Liu et al., 2015; Tian et al., 2013). Lataye et al. (2006) examined the bagasse fly ash from bagasse-fired boilers for the adsorption of pyridine. The maximum removal of pyridine was determined to be 99% at lower concentrations (<50 mg/dm³) and 95% at higher concentrations (600 mg/dm³), using a bagasse fly ash dosage of 25 kg/m³ at room temperature. Aki and Abraham (1999) applied catalytic supercritical water oxidation to treat pyridine, and the addition of a Pt/ γ -Al₂O₃ catalyst was able to increase the reaction rate, and complete conversion of pyridine was achieved at 370°C. Shen et al. (2015) investigated nitrate as an electron acceptor to enhance pyridine biodegradation under anoxic conditions, and demonstrated that *Paracoccus*, *Thiobacillus* and *Paludibacter* were the dominant species in their anoxic degradation system. Yao et al. (2011) used *Lysinibacillus cresolivorans*, a bacterium capable of phenol biodegradation, to study the substrate interactions of m-cresol and pyridine. The study showed that *L. cresolivorans* had good potential for degrading mixed substrates of m-cresol and pyridine. Li et al. (2009) found that a novel bacterium *Streptomyces* sp. HJ02 could grow using pyridine as its only carbon resource and had strong ability for pyridine degradation. The results suggested that the metabolism of pyridine involved cleaving the molecule between C-2 and C-3, and pyridine was hydrolyzed to succinate semialdehyde and formamide. Because of the strong toxicity of pyridine toward microbes, a high processing cost and long processing cycle are required to obtain satisfactory results by routine biological treatments. In addition, it was shown that the inhibition of microbial growth by nitrogen heterocyclic compounds can be weakened through destroying the ring structure (Zhang et al., 2014). Thus, developing a new type of treatment technology as a pre-treatment or deep treatment to destroy the structure of heterocyclic nitrogen compounds in drinking water treatment would have great significance.

The dielectric barrier discharge (DBD) apparatus is based on the use of a discharge electrode plate covered with a dielectric barrier of glass, quartz, ceramics, mica or alumina, etc., which can uniformly distribute the discharge over the entire electrode area. To prevent spark formation and eliminate electrode corrosion, the dielectric has strict requirements. A high frequency voltage is input between the plates so that a micro-discharge of filament-shaped pulses can be formed to excite background molecules and produce large quantities of active species such as ·OH, O₃, O²⁺ and so on (Jiang et al., 2014; Hijosa-Valsero et al., 2013). These active species have strong activity and can induce chain reactions in pollutant molecules. As a result of its strong oxidizing capability, complex organic pollutants in drinking water could be degraded fully into CO₂ and H₂O, conforming to the principles of green oxidation technology (Wang et al., 2014). Compared with other discharge types such as corona discharge (Wan et al., 2011), glow discharge (Liu et al., 2016), microwave discharge (Shiyan et al., 2016) etc., the dielectric barrier strong ionization discharge can form a uniform, diffuse and stable

discharge. The electric field intensity between the discharge plates is greater than 380 Td and even as high as 800 Td or more, and the electron average energy can reach more than 10 eV (Bai et al., 2005). Based on the theory of strong ionization discharge and the oxidizing properties of active chemical species, a strong ionization discharge system for use in drinking water treatment was developed in this study. Strong ionization discharge technology uses O₂ and H₂O as materials and generates no pollution, in accordance with the green treatment concept for environmental pollution (Zhang et al., 2002). Compared with other treatment processes, the technology has the advantages of short processing cycle, high processing efficiency, smaller footprint, no secondary pollution, etc. This work adopted dielectric barrier strong ionization discharge technology that was able to overcome the shortcomings of the existing technology, such as small water treatment volume due to the restrictions of plate clearance, discharge medium and energy consumption. This system can achieve 2 t/h water treatment volumes through improvements in structure and process parameters. The degradation characteristics of pyridine by DBD were researched in this study, and the results provide a valuable reference for the efficient treatment of nitrogen heterocyclic compounds in drinking water.

1. Experiments

1.1. Experimental apparatus

The experimental device mainly consisted of a generator producing active species by dielectric barrier strong ionization discharge, and a water circulation system. The generator operates by dielectric barrier strong ionization discharge technology. Using a high voltage high frequency power supply capable of producing high voltage/high frequency of 2.5–4 kV/15–23 kHz to input between the discharge electrode and the ground electrode, the O₂ molecules were strongly ionized to produce more active species such as O, O₃, O₂⁺, and O₂⁻. The discharge electrodes were made of sintered silver metal, and their outer surface was sprayed with a thin (330 μm) dielectric layer composed of dense insulating α -Al₂O₃ powder. A stainless steel plate was processed to form a cavity through which water or air could flow to regulate the temperature of the plasma reaction process and used as the ground electrode, as shown in Fig. 1. The structure of the discharge area was made up of three hollow grounding plates (plate size 19.5 cm × 10 cm × 1 cm) and two groups of discharge electrodes (plate size 19.5 cm × 10 cm × 1 mm). A grounding plate was placed in the bottom, with a discharge plate placed above it, and so on. Insulating tapes were placed between the discharge plate and the grounding plate to maintain plate clearance of 0.2 mm. After completion of assembly, glass cement was used to seal the shell to avoid leakage of O₂. Due to the outstanding characteristics of the dielectric, a large number of stable, uniform, micro-fine-filament pulse discharges could be formed in the discharge gap. The discharge intensity of electric field in the discharge gap can be described as follows (Zhang et al., 2013):

$$E_g = \frac{V\varepsilon_d}{2l_d\varepsilon_g + l_g\varepsilon_d'} \quad (1)$$

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