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Ozonation of endocrine disrupting chemical BHA under the suppression effect by salt additive—With and without H_2O_2

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Abstract

The oxidation of fresh and saline wastewater containing an endocrine disrupting chemical (butylated hydroxyanisole, BHA) under different reaction conditions by ozonation and O_3/H_2O_2 was investigated at various pH levels. The observed pseudo-first-order reaction kinetics was justified through a combined direct ozone and indirect radical oxidation approach for the ozonation process. The BHA decay rates increased with the increase of the solution pH, but decreased as the NaCl concentration increased because of the consumption of ozone by chloride. A kinetic model was therefore derived for predicting BHA degradation at various initial pH levels and NaCl concentrations. For the O_3/H_2O_2 and $O_3/H_2O_2/Cl^-$ processes, the rate of BHA removal was investigated at hydrogen peroxide concentration ranged from 0.5 to 5 mM at pH 7. Different optimal H_2O_2 dosages and decay rates were found for both processes due to the participation of reactions among O_3, H_2O_2, $OH^\bullet$ and Cl^- as discussed in the paper.
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1. Introduction

Butylated hydroxyanisole (BHA) is recognized as an endocrine disruptor chemical (EDC) in animal experiments [1–3]. It is a synthetic antioxidant which has been widely used to preserve and stabilize the freshness, nutrition, flavour and colour of food and animal feed products [4]. It was also added to edible fats and fat-containing food as an antioxidant, which prevents food from becoming rancid and developing objectionable odours. However, its use is not permitted in some countries such as Japan, because BHA could be tumour promoter [5]. Recently, numerous studies have shown the carcinogenicity of BHA in rat and hamster fore stomach, disturbance in mitochondrial electron transport, slightly oestrogenic to breast cancer cells, binds rainbow trout oestrogen receptor and claimed to stimulate transcriptional activity of the human oestrogen receptor [4,6].

In this study, BHA is selected as the probe for the investigation of the treatability by an advanced oxidation process (AOP): O_3/H_2O_2 system. Karimi et al. [7] has defined the AOP as processes which involved the generation of hydroxyl radicals in sufficient quantity, such as O_3/OH^-, O_3/H_2O_2, Fe^{2+}/H_2O_2, UVC/H_2O_2, UVC/O_3 and UVA/TiO_2 [8]. Kamenev et al. [9] investigated the effect of H_2O_2 concentration, $[H_2O_2]$, on the removal of phenol in O_3/H_2O_2 process and did not observe any significant impact on the oxidation efficiency for H_2O_2 doses ranging between 0.29 and 0.88 mM. It is postulated that these concentrations were relatively high comparing to the low concentrations of phenol (0.04 mg/L or 0.42 μM). Therefore, the effects of H_2O_2 could not be observed. On the other hand, Gulyas et al. [10] reported that the significant influence of H_2O_2 concentration on the removal of triethylene glycol dimethylether during O_3/H_2O_2 oxidation process. Therefore an optimum dosage of H_2O_2 towards ozonation is necessary for investigation.

Sodium chloride is commonly used as retarding agents or exhausting agents in textile industries with a concentration ranging from 5 to 100 ppt (part per thousand salinity or g/L) [11,12]. In Hong Kong, seawater is used for flushing purpose. The wastewater received by treatment works is usually salty (salinity of seawater is about 30 ppt) with an average pH level of 8 [13]. The chloride ions in wastewater may exert

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interference toward the performance of ozonation either in acidic or alkaline conditions. Muthukumar and Selvakumar [14] reported that presence of salts content in the acid dye effluent increase the complete decolouration time and hence decrease the decolouration efficiency of ozone. Therefore, the presence of NaCl in the treatment was simulated in this study as well.

2. Materials and methods

The butylated hydroxyanisole (BHA) was obtained from Sigma–Aldrich® (98.1% Purity, CAS: 25013-16-5). Sodium chloride and hydrogen peroxide (30%, w/w) were obtained from VWR International Ltd. and Junsei Chemical Co. Ltd. (Japan), respectively. All the solutions were prepared by 18 MΩ deionized distilled water from a Bamstead NANO pure water treatment system. All chemicals and solvents are of HPLC grade, and they are used without further purification. For pH adjustment, 0.1 M sulphuric acid or 0.1 M sodium hydroxide was used. All experiments were carried out at a controlled room temperature at 23 ± 2 °C.

For the ozonation process, the experiments were carried out in a 1000 ml cylindrical contact tower, where the deionized water solution was pre-ozonated for 10 min (to reach saturation of ozone). Ozone gas was fed continuously into the reactor throughout the reaction with a glass sparger for better gas transfer and mixing. The OZAT® ozone generator (CFS-1A from Ozonia Ltd.) using oxygen as the source generated 0.018 mM (0.88 mg/L) saturated ozone at a flowrate of 1 L min^{-1}. The dissolved ozone concentration was determined by Indigo spectrometric method.

Sample analysis by HPLC was carried out using a chromatograph equipped with a pump (Waters 515), an ISCO injector (with a 20 μl loop), a C8, 250 mm × 4.6 mm, 5 μm particle size Restek® Pinnacle II column, and a Waters™ 486 Tunable Absorbance Detector. The mobile phase was 70% (v/v) acetonitrile and 30% distilled-deionised water, and the flow rate was 1.0 ml/min. The detection wavelength for BHA was 226 nm, which were pre-determined as the maximum absorption wavelengths (λ_{max}) by a Spectronic Genesys 2™ UV–vis Spectrophotometer.

In each test, 0.2 mM of BHA (36 mg/L) in a continuous ozone purging mode was applied to investigate the reaction kinetics of BHA ozonation (i.e. 0.024 mg O_3/mg BHA^{-1}). Three initial pH levels at 3, 7, and 11 were used to examine the consequence of the initial pH levels to the overall reaction rates. Samples were collected from the base of the contact tower at specific time intervals (0, 1.5, 3, 5, 8, 10 and 20 min) and reaction was quenched by excess sodium thiosulphate [15,16]. For those experiments involving H_2O_2, different concentration of H_2O_2 (0.5–5 mM) has been spiked simultaneously before the reaction start, in order to quench the residual hydrogen peroxide in the sample, excessive methanol [17] is previously added into the vials in a 1:1 ratio. Experiments involving the salt were carried out by addition of 10, 20, 30 ppt NaCl to the pre-ozonated solution and followed by the introduction of BHA.

3. Results and discussion

3.1. Quantitative prediction: decay kinetics of BHA under different conditions

The ozonation of BHA at different initial pH levels and NaCl concentration was examined. More than 98% of 0.2 mM BHA could be removed by continuous feeding of ozone at 0.018 mM in 20 min when no NaCl was present in the solution, where the decay rates of BHA were found to follow pseudo-first-order kinetics (Fig. 1). In the presence of NaCl, the interfering effect causing the drop of ozonation efficiency was observed at three different salinities (10, 20, and 30 ppt NaCl), and Fig. 2 (solid lines) shown one of the typical example (i.e. 30 ppt). The kinetic rate constants of ozonation reduced by 10%, 13% and 16% after the addition of 10, 20, and 30 ppt of NaCl, respectively. Therefore, it should be useful to establish a model that predicted the suppression effect for the proper reactor design when

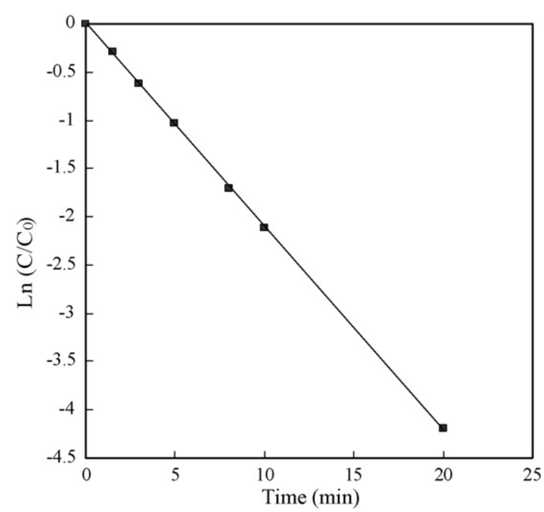

Fig. 1. Ozonation of 0.2 mM BHA at pH 7 in a pseudo-first-order kinetics.

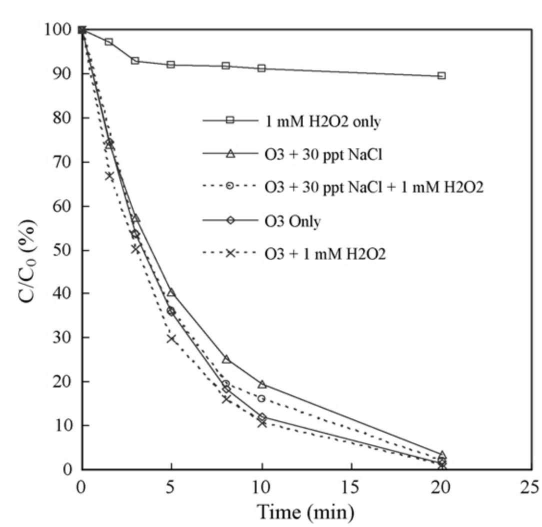

Fig. 2. Removal efficiency of 0.2 mM BHA by different ozonation conditions at pH level of 7.

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