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Evaluation of biodegradability and oxidation degree of hospital wastewater using photo-Fenton process as the pretreatment method

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Abstract

In this work, the photo-Fenton process was used for the pretreatment of hospital wastewater with the objective of improving its overall biodegradability and determining the degree of increased oxidation. The chemical oxygen demand (COD), 5-day biochemical oxygen demand (BOD_5), total organic carbon (TOC) and toxicity towards the gram negative marine bioluminescent bacteria of the species *V. fischeri* were selected as the environmental sum parameters to follow the performance of this process. The enhancement of biodegradability, evaluated in terms of the BOD_5/COD ratio, increased from 0.3 to 0.52 and the oxidation degree, calculated in terms of AOS, leveled up from −1.14 to +1.58 at the optimum conditions; a dosage ratio of COD:H_2O_2:Fe(II) at 1:4:0.1, and a reaction pH of 3. The reduction in the inhibition percentage from the toxicity test indicated the safe levels for micro-organisms in degrading the residual organic substance in this method. Almost total removal percentages of COD, BOD_5, and TOC were found by a sequential activated sludge process for the pre-treated wastewater. Results obtained from this work indicated that the photo-Fenton process could be a suitable pretreatment method in reducing toxicity of pollutants and enhancing biodegradability of hospital wastewaters treated in a coupled photochemical–biological system.

Keywords: Biodegradation; Hospital wastewater; Chemical oxidation; Activated sludge; Aerobic process; Photo-Fenton

1. Introduction

Hospital wastewater contains a variety of toxic organic substances such as pharmaceuticals, radionuclides, solvents, and disinfectants for medical purposes in a wide range of concentrations due to laboratory and research activities or medicine excretion [1–5]. The contact of hospital pollutants with aquatic ecosystems leads to a risk directly related to the existence of hazardous substances, which could have potential negative effects on the biological balance of natural environments [4]. Pollution from hospital wastewaters can have effects even at low concentrations. Aquatic organisms, for instance, respond negatively to low concentrations of formaldehyde, which is a frequently found contaminant in hospital wastewater [6]. It was reported that formaldehyde in the range of 10–100 mg/L was toxic to the microbial in wastewater treatment system [7–9]. In addition, the presence of organochlorine compounds in high concentrations in hospital effluent has also been reported as toxic to aquatic life [10]. The halogenated organic compounds adsorbable on activated carbon (AOX) up to 10 mg/L were found in hospital wastewater [11].

In general, these toxic chemicals in hospital wastewater were discharged directly to wastewater treatment plants, which mostly employ biological treatment process. The failure of the wastewater treatment plant according to adverse effects of the contaminants on the community of organisms in charge of the biological decomposition of the organic matter was reported [11]. Attempts to reduce the toxicity of the chemicals in hospital were focused in many previous works [12–14]. The toxicity of formaldehyde in hospital wastewater was successfully reduced by photo-Fenton process [12]. Reverse osmosis, activated carbon, and ozonation have been shown to significantly reduce or eliminate antibiotics and pharmaceutical substances presented in the wastewater [13,14]. A submerged hollow fiber membrane bioreactor (MBR) was also proposed in treating the hospital wastewater [15]. As many treatment methods exhibit the possibility in remove the

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toxic species from wastewater effluent; however, most wastewater treatment facilities do not employ these techniques. To provide the alternative method in treating hospital wastewater, the pretreatment process to reduce the toxicity of pollutants and enhance biodegradability of the wastewater prior to discharge the waste stream to the existing biological treatment plant is proposed in this work.

The pretreatment process in this work is photo-Fenton, an emerging and very promising technology based on the oxidation of hazardous organic compounds in several wastewater types [16–18]. This process involves the use of one or more oxidizing agents, usually hydrogen peroxide and/or oxygen, and a catalyst, a metal salt or oxide (usually iron) [19–21]. This method is attractive due to the abundance and non-toxicity of iron. As reported by many previous works, the Fenton process in its unmodified form is efficient only in the acidic range and is usually most efficient at around pH 2.8 [22]. Uses of Fenton process can lead to the complete mineralization of some organic compounds, converting them to CO_2, H_2O and inorganic ions. However, the mechanism of Fenton's reactions is not yet completely cleared [21,23,24].

The prime objective of this study is to evaluate the improvement in biodegradability and oxidation degree of pollutants in hospital wastewater using the photo-Fenton process. The results are evaluated as both BOD_5/COD ratio and the increasing of the oxidation degree in terms of AOS. Additional information in toxicity reduction is also provided in this paper. The appropriate conditions for the photo-Fenton process for application to hospital wastewater are also reported for the design of the treatment process.

2. Material and methods

2.1. Wastewater source and characterization

The studied wastewater was obtained from one hospital in Bangkok, Thailand. The total volume of wastewater of this hospital is estimated at 350 m^3/day. Over a time-period of 28 days, the 24 h composite wastewater samples were taken daily due to the large variations in concentration between the different departments. The studied wastewater in this work was from analysis room (60%) and laboratory (40%). The wastewater from embalming room is not included in this work. The wastewater samples were collected from each source before entering into the entire hospital sewer network, which discharged the effluents into the biological wastewater treatment plant without pretreatment. The wastewater treatment plant failed to function for several months due to the discharging of toxic effluents from hospital activities. All the water samples were kept at 4 °C until analysis or experiments.

All reagents used in this experiment were all analytical grade and used as received without further purification. Hydrogen peroxide solution (35%, w/w), heptahydrated ferrous sulfate ($FeSO_4{\cdot}7H_2O$), and NaOH were purchased from Merck (Germany). Water used throughout was prepared with Millipore Milli-Q water.

2.2. Experimental method

The experimental apparatus consisted of a cylindrical quartz reactor (7841-06, Ace Glass; Vineland, NJ), a double-walled quartz cooling water jacket, and a 10 W germicide lamp with a nominal wavelength range of 254 nm. The cooling water jacket was set up inside the reactor to maintain the temperature to be within a range of 25–31 °C, preventing excessive heating of the reaction. A lamp was placed inside the cooling water jacket. The volume of aqueous solution in all conducted experiments was 1000 mL. The reaction solution was stirred with a magnetic stirrer using a constant speed at 150 rpm to maintain a well-mixed solution during the experiments. Before turning on the UV lamp, the wastewater was placed in the dark, covered with aluminum foil, and the solution was adjusted to the desired pH. The addition of adequate amounts of photocatalysts (H_2O_2 or Fe^{2+}/H_2O_2) was added to the hospital wastewater only at the beginning of irradiation process in the batch mode. Samples were retrieved from the reactor for analysis at different time intervals. Photo-Fenton was stopped instantly by adding NaOH to the reaction mixtures and quenched by adding Na_2SO_3 before analysis. The reaction period in irradiation process was 2 h for all experiments.

Changes in chemical oxygen demand (COD) were determined by means of the dichromate reflux standard method [25], biological oxygen demand (BOD_5) and pH were measured by procedures described in standard methods [25]. Soluble COD (SCOD) and soluble BOD (SBOD) were measured after filtration (0.45 mm filter paper), and used to evaluate the treatment efficiency of biological treatment process after photo-Fenton pretreatment. Initial and treated total organic carbons (TOC) were analyzed with a Shimadzu 700 TOC ANALYZER 0-1 Analytical after filtration. Hydrogen peroxide was measured by the standard iodometric titration method [26]. Experiments were done in duplicate for the same set of conditions. The variations were systematically within ±10% of the stated values.

2.3. Toxicity test procedures

The bioassay on bacteria luminescence was carried out with a LUMIStox system (Dr. Lange GmbH, Duesseldorf, Germany) following the procedure of European standard NFEN ISO 11348-3. Tests were performed using gram negative marine bioluminescent bacteria of the species *V. fischeri* NRRL-B-11177 of the *Vibrionaceae* family. Samples were filtered using a 0.45 μm pore size membrane to prevent TSS interferences on bacteria luminescence. The samples were treated with NaCl solution of 20 g/L and brought to 50 mS/cm conductivity before analysis. Starting from the concentration of the sample, eight consecutive dilutions were tested (dilution factor 1:2); the inhibition of bioluminescence was measured at a wavelength of 490 nm, with readings after 5 and 15 min of incubation at 15 °C. The EC_{50} values were calculated as reported by Bulich [27].

2.4. Biological procedure

To obtain the biodegradability information, the activated sludge system was applied in cylindrical aeration glass-vessels