



Research article

Degradation of organic pollutants by photoelectro-peroxone/ZVI process: Synergistic, kinetic and feasibility studies

Mehdi Ahmadi^{a,b}, Farshid Ghanbari^{c,*}^a Environmental Technologies Research Center, Ahvaz Jundishapur University of Medical Sciences, Ahvaz, Iran^b Department of Environmental Health Engineering, School of Public Health, Ahvaz Jundishapur University of Medical Sciences, Ahvaz, Iran^c Department of Environmental Health Engineering, Abadan School of Medical Sciences, Abadan, Iran

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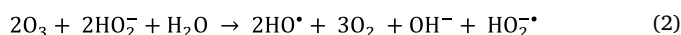
ABSTRACT

In this study a novel hybrid process was employed for the degradation of Metanil Yellow (MY). The operational parameters of photoelectro-peroxone/zero valent iron (PEP/ZVI) process were studied and the complete decoloration was found at pH = 3.0, 100 mg/L ZVI, 33.2 mg/L ozone, 300 mA applied current and 25 min reaction time. The combination of UV, ozone, electrogenerated H₂O₂ and ZVI showed high synergistic effect for MY degradation. ZVI showed high reusability in PEP/ZVI process. Among anions, nitrite ion demonstrated high inhibitory effect while chloride ions had no significant effect on MY degradation. Scavenging tests depicted that hydroxyl radical and singlet oxygen were the main agents of MY degradation. PEP/ZVI process was tested for several emerging pollutants (benzotriazole, 4-chlorophenol, carmoisine and tetracycline); the results presented the effectiveness of the process for the degradation of pollutants in a way that complete degradation occurred at only 30 min. Moreover, the performance of PEP/ZVI was examined for the treatment of two actual wastewaters. PEP/ZVI demonstrated an excellent function in terms of the removal of organic compounds to achieve discharging standards.

1. Introduction

Various processes have been suggested for the degradation of organic pollutants in literature. Based on destructive processes, advanced oxidation processes (AOPs) have been successfully examined for the degradation of synthetic dye solutions and the treatment of actual dying wastewaters (Asghar et al., 2015; Munoz et al., 2015). AOPs are usually defined as the oxidation of organic compounds by means of highly reactive species especially hydroxyl radicals (HO[•]) with redox potential of 2.7 V (Pignatello et al., 2006). Fenton oxidation (H₂O₂/Fe²⁺), direct and catalytic ozonation, photocatalysis, electrochemical processes are conventional AOPs which have been extensively employed for water and wastewater treatment (Ahmadi et al., 2016, 2017; Gutierrez-Mata et al., 2017). Hybrid advanced oxidation processes recently received lots of attention from researchers for the acceleration of degrading organic pollutants. Accordingly, two or more processes are combined to increase the degradation rate of pollutants (Chakma et al., 2015). Numerous studies have been increasing for the application of hybrid AOPs to depollute wastewater and contaminated water including electro-Fenton, electro-photocatalysis, sono-photo-Fenton, etc (Mahamuni and Adewuyi, 2010). In recent years, electro-peroxone (EP) has been used

for the removal of persistent organic pollutants. EP comprises electro-generation of hydrogen peroxide and ozonation in an electrochemical cell (Turkay et al., 2017). In fact, peroxone reagent (H₂O₂/O₃) was electrochemically formed in electrochemical reactor. H₂O₂ is generated through the reduction of oxygen at the carbon-based cathodes while ozone is sparged into the solution. Free radicals are produced through following reactions (Fischbacher et al., 2013; Frangos et al., 2016).



Decomposition of ozone at the cathode also increases the rate of hydroxyl radical generation within EP reactor (Jaafarzadeh et al., 2017; Wang et al., 2018).

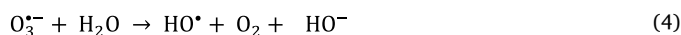


Photo-assisted EP has been used to enhance the activation of ozone and hydrogen peroxide. In fact, photoelectro-peroxone (PEP) is a hybrid process consisting of ozonation, electrolysis and UV irradiation (Jaafarzadeh et al., 2017; Shen et al., 2017).

* Corresponding author.

E-mail address: Ghanbari.env@gmail.com (F. Ghanbari).



Ferrous ion can be also added to PEP reactor for the generation of Fenton reagent. Jaafarzadeh et al. demonstrated that adding ferrous and copper ions enhanced the degradation rate of 2,4-dichlorophenoxyacetic acid (Jaafarzadeh et al., 2017). While another study showed that ferrous ions had an inhibitory effect on total organic carbon (TOC) removal in EP process (Frangos et al., 2016).

Zero valent iron (ZVI) has attracted the interest of environmental scientists for pollutant removals thanks to its high performance, low cost and environmentally friendly matter (Yazdanbakhsh et al., 2015). ZVI is a strong reductant which is extensively applied in groundwater remediation. In this way, ZVI is also used for the activation of chemical oxidants to generate free radicals (Yamaguchi et al., 2018). ZVI as the source of Fe^{2+} can be added to PEP process to improve the function of system through the formation of Fenton reagent (Frangos et al., 2016; Jaafarzadeh et al., 2017).

To the best of our knowledge, PEP has been recently introduced to wastewater engineers for the depollution of contaminants. Therefore, there are few studies related to pollutants removal from aqueous solutions using PEP. Moreover, no study has been conducted on PEP process in the presence of ZVI as catalyst (PEP/ZVI). Most of researches have been focused on synthetic solutions to evaluate the system. Therefore, there is a few information about the function of PEP process in actual wastewater treatment.

In this work, PEP/ZVI was used for metanil yellow (MY) degradation as a sample of organic pollutants. The effects of applied current, ozone dosage, ZVI dosage and pH were studied and then the kinetic of MY degradation was investigated. The mechanism of MY degradation was demonstrated by scavenging experiments. The performance of PEP/ZVI was tested for the degradation of some organic pollutants (benzotriazole, carmoisine, tetracycline, 4-chlorophenol). For the first time, PEP/ZVI process was performed on the actual textile and pulp and paper wastewaters.

2. Materials and methods

2.1. Chemicals

All chemicals used in this study were in analytical grade and all solutions were prepared with deionized water. Metanil yellow ($\text{C}_{18}\text{H}_{14}\text{N}_3\text{NaO}_3\text{S}$) and carmoisine ($\text{C}_{20}\text{H}_{12}\text{N}_2\text{Na}_2\text{O}_7\text{S}_2$) with purity of 99% were purchased from Alvan Sabet (Iran). Zero valent iron (ZVI) was provided from Merck Company with size less than 150 μm . Sodium sulfate (Na_2SO_4), sodium nitrate, sodium nitrite and sodium chloride were purchased from BDH Company. Ethanol, *tert*-butyl alcohol, sulfuric acid, *para*-benzoquinone (p-BQ), L-histidine, tetracycline (HPLC grade) and ferrous sulfate were purchased from Sigma-Aldrich. Benzotriazole (98%) and 4-chlorophenol (98%) were provided from Merck Company. Acetonitrile (HPLC grade), methanol (HPLC grade) and water (HPLC grade) were purchased from Samchun Inc.

2.2. PEP/ZVI process

PEP reactor used in this work was a quartz cylinder (6.0 cm diameter and 16.1 cm height) as an undivided cell. The experiments were carried out at the temperature of 25–28 °C. MY solution (300 mL, 50 mg/L concentration) was prepared in an electrolyte (0.01 M Na_2SO_4). A graphite felt (70 mm \times 95 mm \times 3 mm) (Sazeh Morakab Inc., Iran) was used as cathode. A platinum (Pt) sheet (99%) (10 mm \times 20 mm) was applied as the anode. The distance between the electrodes was 3.0 cm. A mixture of ozone and oxygen was sparged by an ozone generator device (Arda, A121) via a ceramic diffuser. Ozone concentrations was adjusted using changing the power of ozone generator. The flow rate was constant for the mixture in all conditions (0.25 L/min). To apply electrical current, a digital DC power supply

(3.000 A, 30 V) was employed under galvanostatic conditions. Two low pressure UVC lamps (4 W, Philips) were placed out of the reactor with maximum emission at 254 nm wavelength. The lamps were posited at the both sides of the reactor. The space of 2 cm was considered between the UVC lamps and electrochemical cell. UV intensity was determined by a radio meter (Lux-UVIR meter, Leybold Didactic GMBH-666-230) and was found as 1.51 mW/cm^2 at the center of reactor. The solution was mechanically stirred to provide sufficient mixing in the electrochemical cell. UV lamp was turn on for 10 min before reaction time to warm up. Ozone generator and DC power supply were simultaneously turned on to conduct PEP. A known amount of ZVI was added to the solution. The samples were withdrawn at each 5 min. All experiments were conducted at triplicate and mean values were reported for results. Standard deviation was less than 2.7% for the results of MY removal. Other processes (electrogeneration of H_2O_2 (EH), UV-EH, ozonation, UV/ozonation, EP, ZVI, EH/ZVI, UV/ZVI, UV/ozonation/ZVI, ozonation/ZVI and PEP) were conducted at the same conditions. PEP/ZVI was evaluated for the treatment of actual wastewater. Pulp and paper wastewater and textile wastewater were collected from Khuzestan province (Iran) and Markazi province (Iran) respectively. The samples were stored in commercial refrigerator at 4 °C.

2.3. Analytical methods

Metanil yellow was measured by a spectrophotometer (DR-6000) in visible region at the wavelength of 436 nm as absorbance the maximum of metanil yellow. The total organic carbon (TOC) of the solution was measured by TOC ampules in low range (Merck). The accuracy of TOC values was controlled by potassium hydrogen phthalate (KHP) based on the Standard Methods. Chemical oxygen demand (COD), biochemical oxygen demand (BOD), iron, color, sulfate, chloride, bicarbonate ions and total dissolved solids (TDS) were determined according to the Standard Methods (APHA, 1999). The measurement of benzotriazole, 4-chlorophenol, carmoisine and tetracycline were presented in supplementary information. Ozone and hydrogen peroxide concentrations were measured by iodometric method; oxidants oxidize iodide to iodine in the presence of acid. Iodine is titrated by thiosulfate solution in the presence of starch (Vogel, 2013).

3. Results and discussion

3.1. The effects of pH, applied current, ozone dosage and ZVI

The effect of pH was investigated under the conditions of 200 mA applied current, ozone dosage of 24.3 mg/L and 100 mg/L ZVI. Fig. 1a shows the decoloration of MY in different pH values. As can be seen, the maximum efficiency was found at pH = 3.0 (87.2%). In pH values of 2–9, the wavelength of maximum absorbance of MY did not change. Hence we conducted experiments in this range. In pH < 2, the color of MY solution was changed to red color. Increasing in pH value suppressed the removal efficiency. Acidic conditions are favorable media for the electrogeneration of H_2O_2 based on Eq. (1). ZVI releases ferrous ions at acidic pH; at pH > 4.5, ferrous ions were precipitated in form of $\text{Fe}(\text{OH})_n$ (Şahinkaya, 2013). These hydroxides are not able to activate hydrogen peroxide for the generation of hydroxyl radical. Moreover, the best pH for Fenton oxidation is well known in the range of 2–4. At alkaline conditions, electrogenerated H_2O_2 was self-decomposed to oxygen and water. According to the results, pH was an important parameter in this system since directly or indirectly affected ferrous species, electrogeneration of H_2O_2 and amount of H_2O_2 (Weng et al., 2013).

The effect of applied current was investigated in range of 100–400 mA under the conditions of pH = 3.0, ozone dosage of 24.3 mg/L and 100 mg/L ZVI (Fig. 1b). With increasing of applied current, MY removal was enhanced in which decoloration efficiencies were 70, 84.2, 100 and 100% for 100, 200, 300 and 400 mA

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