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Treatment of organic pollutants in coke plant wastewater by the method of ultrasonic irradiation, catalytic oxidation and activated sludge

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Abstract

The paper deals with the degradation of the organic pollutants in coke plant wastewater by the combination process of ultrasonic irradiation, catalytic oxidation and activated sludge. The effect factors of ultrasonic irradiation on the degradation of the organic pollutants such as saturating gas, initial pollutant concentration, ultrasonic power density, the category and consumption of catalyst were investigated. The results indicate that putting the saturating gas into the reaction solution in the process of the ultrasonic irradiation, low COD initial concentration and high ultrasonic power density are the favorable conditions for their degradation. Compared with single activated-sludge process, co-approach of ultrasonic irradiation and activated-sludge can greatly increase the COD degradation efficiency. When the wastewater was firstly treated by ultrasonic irradiation process and then followed by activated-sludge process for 240 min, respectively, the COD degradation efficiency increased by 48.29–80.54%. Additionally, when 3.0 mmol/l of ferrous sulfate was added into the ultrasonic irradiation process, the COD degradation efficiency was as high as 95.74%, 63.49% higher than that of the activated-sludge approach alone.

Keywords: Coking wastewater; COD degradation; Combination process; Water quality model

1. Introduction

The coke plant wastewater consists of many kinds of components, which are usually refractory aromatic organic compounds such as naphthalene, quinoline, pyridine and so on. At present, the activated sludge process is usually treated coke plant wastewater. With the activated sludge process, the outlet water usually contains about 350–700 mg/l COD, which cannot meet the demand of the drainage by environmental protection. Because some organic compounds in the coke plant wastewater are toxically refractory and inert to the activated sludge process, the process of activated sludge alone cannot decompose the organic pollutants in coke plant wastewater efficiently. So, it is very important to find a primary treatment process to decompose these toxically refractory and inert organic compounds firstly.

The research on chemical ultrasonic began in 1927. Richards and Loomis [1] reported the acceleration of conventional reactions and the reduction-oxidation process by ultrasound. Since then, a number of chemical reactions have been observed in an ultrasonic field [2–4]. In recent years, attention has focused on the application of ultrasonic energy to solve the problems associated with water pollution, especially in removing toxic and hazardous organic compounds from contaminated water [5,6]. For many contaminants, the ultrasonic process has the advantage of completely destroying or converting these organics, not simply transferring them to another medium.

The chemical effects of ultrasound are depended on the phenomenon of acoustic cavitation [7]. Sound is transmitted through any fluid as a wave consisting of alternating compressing and rarefaction cycles. If the rarefaction wave is sufficiently powerful, it can produce a negative pressure, which is large enough to overcome the intermolecular forces

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Nomenclature

A	activated sludge process
C	catalytic oxidation process
CP	combined process
COD	chemical oxygen consumption (mg/l)
EB	easily biodegradable COD (mg/l)
HB	hardly biodegradable COD (mg/l)
I	inlet
NB	inert COD (mg/l)
OR	oxygen consumption rate (mg/(l min)
RC	removed COD (mg/l)
S	area for dissolved oxygen in Fig. 12 (mg/l)
U	ultrasonic irradiation process
YH	cell production rate coefficient

binding the fluid. As a result, the molecules are torn apart from each other and form tiny micro bubbles. These micro bubbles gradually grow during the compression–rarefaction cycles until they reach a critical size. Subsequent compression then causes the micro bubbles to collapse almost instantaneously, thereby releasing a large amount of energy. Temperatures of the order of 5000 K have been experimentally obtained [8], and pressures of the order of 1000 atmospheres have been calculated [9].

Primary chemical reactions result in the transient state of these high pressures and temperatures, both during and immediately after collapse of the micro bubbles [10,11]. Solvent and solute vapors in the cavity undergo direct thermal dissociation to yield CO_2, H_2O, and radicals, such as hydroxyl and atomic hydrogen. Some of these radicals may recombine to form new compounds. In the bulk liquid phase, secondary reactions between solute molecules and radicals generated in the cavities also take place. These two reactions scheme are together responsible for the chemical effect of ultrasonic upon the solution. Sometimes, metallic ions act as catalysts to increase the degradation effects of organic pollutants in wastewater by ultrasound [12–14].

In this paper, the results of the degradation effect of the organic compounds in a coke plant wastewater by the ultrasonic irradiation process (UI) at various initial COD concentrations, ultrasonic power density, the action modes of saturating gas, the categories and consumption of catalysts are presented. Combining with the activated sludge process, the ultrasonic irradiation process was developed to decompose the organic compounds in coke plant wastewater completely. The water quality model was used to explain the decomposition effects of different kinds of organic pollutants in the wastewater by the ultrasonic irradiation process.

2. Experimental setup

The experimental apparatus is shown in Fig. 1. The coke plant wastewater came from a Coke Gas Production Plant.

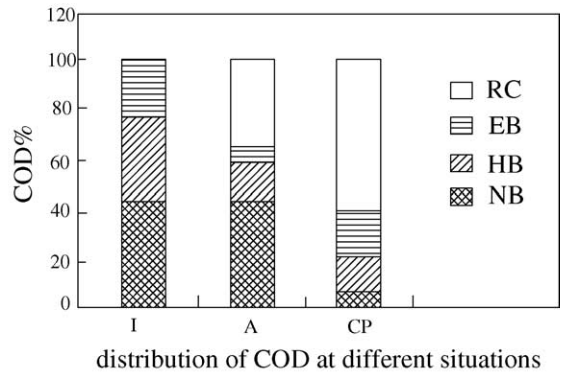

Fig. 1. Equilibrium of COD in the refractory organic industrial wastewater of high concentration. I-inlet or initial COD in waste water: (A) outlet COD in waste water treated by activated sludge process; (CP) outlet COD in waste water treated by combined process; (RC) removed COD; (EB) easily biodegradable COD; (HB) hardly biodegradable COD; (NB) inert COD.

The coke plant wastewater of the 807 mg/l initial COD concentration was prepared. Its initial pH value was adjusted to 8.17. The experiments were carried out at the following standard experimental conditions: initial COD of 807 mg/l, initial pH value of 8.17, reaction time of 240 min. The wastewater was then differently treated with some proportions of sodium hydrogen phosphate, activated sludge or ultrasonic power.

Irradiations of the wastewater were carried out with an ultrasonic generator (made by the Acoustics Research Instate of the China's Science Institute) with an output frequency 18 kHz and power intensity 119.4 kW/m^2. A 500 ml (wastewater volume) glass water-jacketed reactor was equipped with a coarse fritted-glass gas diffuser near the base. A transducer probe of the ultrasonic generator was inserted from the top into the wastewater about 2–3 cm. The temperature of the wastewater was controlled at $25 \pm 0.5\,^{\circ}C$.

The COD concentration was analyzed by potassium dichromate method. EB value means easily biodegradable COD concentration, and HB value means hardly biodegradable COD concentration. While HB value was determined by the difference between the initial value and the sum of EB and NB. The concentration of the dissolved oxygen was determined by a portable multi-parameter measuring meter (the type is Multiline P4) made by WTW Company of Germany.

3. Results and discussion

3.1. Basic principle of the water quality model

The basic principle of the water quality model is shown in Fig. 2. In this mode, the total initial COD in wastewater is divided into three parts: easily biodegradable COD (EB), hardly biodegradable COD (HB) and inert COD (NB). The inert COD is not biodegradable. The concentration of these