



Study of hydrogen gas production coupled with phenol electrochemical oxidation degradation at different stages



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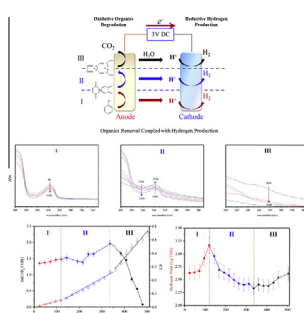
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HIGHLIGHTS

- Protons released from phenol electrochemical oxidation degradation form hydrogen.
- Three stages were observed in phenol electrochemical oxidation degradation at 3 V.
- Each stages could be well described by the first order kinetic model.
- ICE increased in first two stages while decreased dramatically in third stage.
- Hydrogen yield improved in first stage but reduced in second stage.

GRAPHICAL ABSTRACT



ARTICLE INFO

Article history:

Received 4 August 2013

Received in revised form 1 November 2013

Accepted 10 December 2013

Available online 19 December 2013

Keywords:

Hydrogen gas production

Phenol

Electrochemical oxidation degradation

Kinetic

Energy efficiency

ABSTRACT

Hydrogen gas production coupled with phenol electrochemical oxidation was investigated in a novel two-region equipment. The phenol degradation, COD removal, hydrogen production, kinetic, Instantaneous Current Efficiency for COD removal (ICE_{COD}), and hydrogen gas yield (Y_{H_2}) of simulated phenol solution electrochemical oxidation degradation at 3 V applied voltage were surveyed. The results indicated that three stages were observed during the electrochemical oxidation of simulated phenol in this study. The kinetic study showed that first order model well described each stage of phenol electrochemical oxidation process. The ICE_{COD} increased slightly at the initial stage, and dramatically at the second stage, while fell at the finally stage, indicating water molecules were finally participated in the formation of hydrogen due to the degradation of phenol and COD. Hydrogen products from redox reaction of H^+ which release from the degradation of organics oxidation reaction at the anode. Furthermore, the mechanism was discussed and showed that phenol transformed into benzoquinone intermediate in the first step, then the ring was broken and further oxidized into organic compounds like muconic acid, maleic acid or oxalic acid as intermediates in the followed stage, and finally disintegrated into CO_2 and H_2O in electrochemical oxidation process, which demonstrated by the UV and LC-MS images analysis. It could be seen that hydrogen gas production coupled with organic wastewaters electrochemical oxidation would be an effective approach for energy recovery and wastewater reutilization.

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1. Introduction

Phenolic contaminants are a series of the most serious and persistent organic pollutants which widely existed in wastewaters discharged from the chemical plants, coking plants, or petroleum

refineries [1–3]. It is extremely urgent to remove them from the wastewaters before discharging into the environment due to their toxicity, carcinogenicity, teratogenicity and mutagenicity to human beings [1]. However, these wastewaters are usually difficult to be treated just by physical, chemical or biological process [4]. Thus, various of physicochemical approaches, such as coagulation, adsorption, chemical oxidation, have been proposed as the previous or advanced treatments, but these methods are found to

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be ineffective due to the inherent disadvantages in terms of low efficiency, costive, and secondary pollution. Electrochemical oxidation process has been proved to be an effective and environmental friendly technique to treat organic wastewaters because of its easy controlled and environmental compatibility [5]. Furthermore, the coexisted ions contained in the industrial wastewaters and its higher conductivity is beneficial to enhance the organics removal and reduce the cost of electrochemical oxidation process [6,7].

On the other hand, our global energy and environment are suffered from the limitation reserves of fuels and over discharge of greenhouse gases. Hydrogen is considered to be a viable alternative, clean and renewable energy, due to its high energy (122 kJ g^{-1}). The demand of hydrogen gas increases rapidly with a growth rate of nearly 10% per year [8]. Hydrogen gas could be produced through many ways from a broad of raw materials. The predominant means is from fossil fuels; more than 90% of hydrogen was produced by steam reforming of natural gas [9], only about 4% was produced by electrolysis of water owing to the cost for large production [10]. Therefore, exploring effective and clean approaches for hydrogen production coupled with organic wastewater electrochemical oxidation have become one of the greatest challenges for researcher.

Various electrochemical approaches have been proposed to degrade organics in aqueous solutions, such as the degradation of phenol [11,12], dye wastewater [13,14], textile wastewater [15–17], pharmaceutical residues [18,19] et al. Furthermore, electrochemical could be integrated with biological treatment for some wastewaters containing high concentrations of refractory and toxic compounds. Complete mineralization of organics and removal of ammonia were almost achieved in electrochemical oxidation of biologically pretreated coking wastewater [20]. In the investigation of Ahmed Basha et al. indirect oxidation at anode, indirect oxidation by hydrogen peroxide and $\text{UV}/\text{H}_2\text{O}_2$ at the cathode occurred simultaneously in a two-compartment electrolytic cell, separated by an anion exchange membrane, were observed to treat dye effluents [21]. Integrated biological and electrochemical oxidation of pesticide wastewater with high toxicity was investigated [22]. However, in these studies, the hydrogen gas produced in the electrochemical oxidation reactions was not considered.

Moreover, there are some studies conducted to investigate electrolysis hydrogen production coupled with the degradation of organic compounds in aqueous solutions [23,24]. Park et al. demonstrated hydrogen gas production from water splitting by either direct current or solar power coupled with organics oxidation [25–27]. The organic matters and/or NaCl contained in wastewater enhanced the hydrogen production, and colloidal particles seemed beneficial to hydrogen production due to the decomposition of particles and the generation of dissolved organic intermediates [28]. Besides, many electrochemical approaches were demonstrated for hydrogen production from aqueous media contained organics pollutants by different anodes. Juyuan Jiang reported that BDD could be a new electrode for both hydrogen production by cathodic reactions and wastewater treatment by anodic reactions in a single electrochemical cell [29]. However, the membrane in electrochemical systems was polluted by the organic contaminants in the solutions which could reduce the current efficiency and COD removal efficiency. Kargi et al. investigated the hydrogen gas production from waste anaerobic sludge, landfill leachate organics, olive mill wastewater, and industrial wastewater by using different anodes [30–32]. In their investigation, the applied aluminum anode could achieve better hydrogen gas formation and hydrogen energy efficiencies than stainless steel and graphite anodes. However, the soluble activity of these anodes could product secondary sludge contaminants and the consume lots of anode materials. Besides, few works were carried out to investigate phenol and COD removal, hydrogen gas

production, Instantaneous Current Efficiency for COD removal (ICE_{COD}), hydrogen energy efficiency, and hydrogen gas yield (Y_{H_2}) at different time of the whole electrochemical oxidation degradation process.

Therefore, the objectives of this study are to investigate hydrogen gas production coupled with simulated phenol solution electrochemical oxidation degradation in a new electrolyzer system at 3 V applied voltage; COD and phenol removal, hydrogen gas production, ICE_{COD} and COD removal kinetics of anodic reaction, hydrogen energy efficiency of the cathodic reaction, hydrogen yield, and the mechanism of hydrogen gas production coupled with phenol electrochemical oxidation degradation had also studied furthermore.

2. Experiment and materials

2.1. Materials

A new electrochemical oxidation system used in this study was composed of a cylindrical reactor with 500 mL volume, anode, cathode, and DC power supplier (Fig. 1). A cylindrical $\text{Ti}/\text{IrO}_2\text{-RuO}_2$ anode with the diameter of 7 cm and height of 10 cm, a SS cathode with a length of 11 cm and width of 2.5 cm were immersed into 500 mL phenol contained solution. The applied voltage was controlled by a DC power supplier (APS 3005S). The voltage, current, and hydrogen gas produced were recorded every 24 h.

Phenol purchased from Kermel, Tianjin, was used to prepare phenol solution in the electrochemical oxidation experiments. Sodium Sulfate, Sodium Chloride, Sodium Carbonate, Sodium Bicarbonate, Sodium Phosphate, Disodium Hydrogen Phosphate, and Sodium Dihydrogen Phosphate, also supplied by Kermel, Tianjin, were used as the electrolyte to improve the conductivity of the solution. Potassium dichromate, silver sulfate, mercury sulfate, sulfuric acid, ammonium ferrous sulfate, phenanthroline, and ferrous sulfate were used to measure the COD of the solution. Potassium bromide, potassium bromate, natrium carbonicum

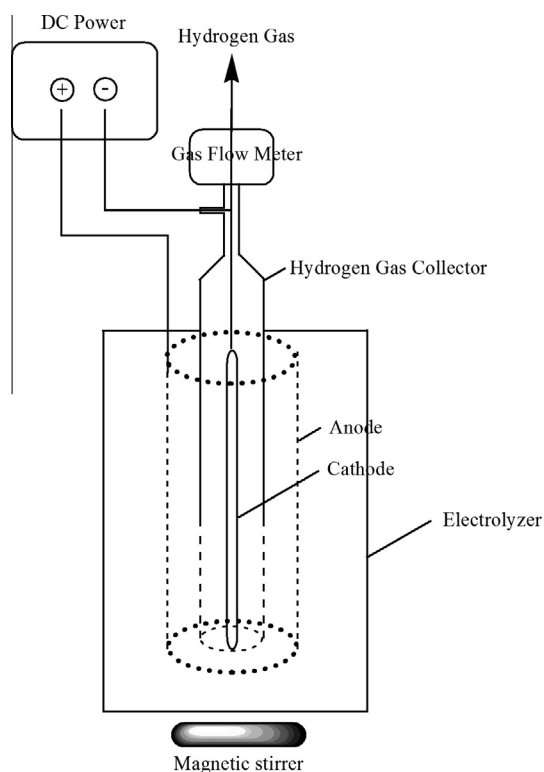


Fig. 1. The new electrochemical oxidation experimental schematic diagram.

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