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An efficient removal of phenol from water by peroxi-electrocoagulation processes

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ABSTRACT

The present study explores the removal of phenol from water by peroxi-electrocoagulation method using mild steel as anode and graphite as cathode. The various parameters like effect of pH, concentration of phenol, current density, temperature, and co-existing ions on the removal efficiency of phenol from the water was studied. These studies were carried out at an initial concentration of phenol of 2.5 mg/L. The results showed that the maximum removal efficiency of 92% was achieved at a current density of 0.10 A/dm^2 and pH of 2.0. The results of pilot scale study show that the process was technologically feasible.

1. Introduction

One of the most widespread forms of organic chemical pollutants in industrial wastewaters is phenol and its compounds. These compounds are very hazardous due to their poor biodegradability, high toxicity and ecological aspects [1]. High concentrations of phenol and its compounds typically are found in aqueous effluents of oil refineries, petrochemical, ceramic, coking and steel plants, coal conversion processes, manufacturers of resin, pharmaceuticals, pesticides, dyes, plastics, explosives and herbicides [1–4]. The presence of phenol in drinking water and irrigation water represents a serious health hazard to humans, animals, plants and microorganisms. Phenol is a potential carcinogen of human, which raises considerable health concerns, even at low concentrations. Its toxic effects include permeabilisation of cellular membranes and cytoplasmic coagulation [6,7]. Phenol is classified by the USEPA as a priority pollutant and represents as one of the most challenging classes of pollutants requiring urgent removal from waste streams. The standard of phenol content in potable and mineral waters is 0.5 μg/L, while the limits for wastewater discharge are 0.5 mg/L for surface waters and 1 mg/L for the sewerage system [8,9]. The most extensively used methods for the removal of phenol from aqueous solutions are adsorption, chemical oxidation, biological treatment, precipitation, distillation, solvent extraction, ion exchange, membrane processes, reverse osmosis, and electrochemical methods [2,4,5,10]. Physical methods like ion exchange, reverse osmosis and membrane processes have proven to be either expensive or inefficient to remove phenol from aqueous solution. At present, chemical treatments are not used due to disadvantages like high costs of maintenance, problems of sludge handling and its disposal, and neutralization of the effluent. Recent research has demonstrated that electrocoagulation (oxidation of sacrificial anode) and electrooxidation methods offer an attractive alternative to above-mentioned traditional methods for treating organic pollutant. The two processes are used either independently or combined in the removal processes.

In present investigation, a combined methodology is followed called "peroxi-electrocoagulation method". This combined process has several distinct advantages. Coagulants introduced without corresponding sulfate or chloride ions are more efficient at removing contaminants from water. By eliminating competing anions and using a highly pure coagulant source, lower metals residuals are obtained and less sludge is produced than when metal salts are utilized. A contaminant free ion source allows maximum removal of the various dissolved forms of pollutant that could be present and require treatment. Apart from the above the presence of $^{\bullet}$OH radical has greater advantage for the removal process. It is reported [11] that as water-containing colloidal particulates, oils or other contaminants move through the applied

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electric field, there may be ionization, electrolysis, hydrolysis and free-radical formation which may alter the physical and chemical properties of water and contaminants [11]. As a result, the reactive and excited state causes contaminants to be released from water and destroyed or made less-soluble [12]. Further, when electrochemical reactors operate at a high cell potential/acidic pH and the anodic process occurs in the potential region of water discharge, hydroxyl radicals ($^{\bullet}OH$) are generated [13–15]. It is also confirmed [15] that ferrous ions act generated in electrocoagulation act as coagulation materials in the electrochemical treatment and catalytically create hydroxyl radicals to a small/larger extent depending upon the conditions.

Following are the reactions taking place in the process,

At anode,

$$2\mathrm{Fe(s)} - 4\mathrm{e}^- \rightarrow 2\mathrm{Fe}^{2+}\mathrm{(aq)} \tag{1}$$

$$2\mathrm{H_2O(l)} - 4\mathrm{e}^- \rightarrow \mathrm{O_2(g)} + 4\mathrm{H}^+\mathrm{(aq)} \tag{2}$$

At cathode,

$$8\mathrm{H}^+\mathrm{(aq)} + 8\mathrm{e}^- \rightarrow 4\mathrm{H_2(g)} \tag{3}$$

$$\mathrm{O_2} + 2\mathrm{H}^+ + 2\mathrm{e}^- \rightarrow \mathrm{H_2O_2} \tag{4}$$

In the bulk,

$$2\mathrm{Fe}^{2+}\mathrm{(aq)} + 4\mathrm{OH}^-\mathrm{(aq)} \rightarrow 2\mathrm{Fe(OH)_2(s)} \tag{5}$$

$$\mathrm{Fe}^{2+}\mathrm{(aq)} + \mathrm{H_2O_2(g)} \rightarrow \mathrm{Fe}^{3+}\mathrm{(aq)} + {}^{\bullet}\mathrm{OH} + \mathrm{OH}^-\mathrm{(aq)} \tag{6}$$

Overall,

$$2\mathrm{Fe(s)} + 5\mathrm{H_2O(l)} \rightarrow 2\mathrm{Fe(OH)_2(s)} + {}^{\bullet}\mathrm{OH} + (5/2)\mathrm{H_2(g)} \tag{7}$$

From the above reactions it is confirmed that the $^{\bullet}OH$ radicals are formed according to reactions (7). To generate $^{\bullet}OH$ radical, the reaction needs H_2O_2 presence in the solution which may be produced following Reaction (4). Further, Reaction (4) needs O_2 presence which may be produced by Reaction (2).

This article presents the results of the studies undertaken on the removal of phenol by peroxi-electrocoagulation method using mild steel as anodes and graphite as cathode. To optimize the maximum removal efficiency of phenol, different parameters like effect of initial phenol concentration, effect of temperature, pH, effect of current density and effect of competing ions like carbonate, phosphate and silicate were studied.

2. Materials and methods

The laboratory electrolytic cell consisted of a 1.0 – L Plexiglas vessel that was fitted with a polycarbonate cell cover with slots to introduce the electrodes, pH sensor, a thermometer and the electrolytes. Mild steel (commercial grade, India) of surface area (0.02 m^2) acted as the anode. The cathodes were graphite (commercial grade, India) sheets of the same size as the anode is placed at an inter-electrode distance of 0.004 m. The temperature of the electrolyte was controlled to the desired value with a variation of ±2 K by adjusting the rate of flow of thermostatically controlled water through an external glass-cooling spiral. A regulated direct current (DC) was supplied from a rectifier (10 A, 0–25 V; Aplab model).

The pilot plant capacity cell (Fig. 1) was designed, fabricated and operated for the removal of phenol from water. The system consists of a DC power supply, an electrochemical reactor, a water tank, a feed pump, a flow control valve, a flow measuring unit, a circulation pump, and treated water tank. The reactor is made of poly-carbonate with an active volume of 1000 L. The electrode sets (anode and cathode) each consist of five pieces of mild steel and graphite sheets, situated approximately 4 mm apart from each other and submerged in the solution. The cell was operated at a current density of 0.1 A/dm^2 and the electrolyte pH of 2.0.

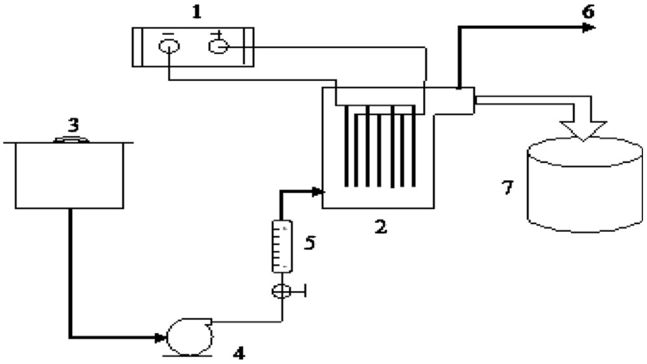

Fig. 1. Flow diagram of the pilot plant electrocoagulation system. 1. DC power supply, 2. Electrocoagulation cell, 3. Water tank, 4. Inlet pump, 5. Flow meter, 6. Gas outlet, 7. Treated water.

The phenol (Analar Reagent, Merck, Germany) was dissolved in deionised water for the required concentration. 0.90 L of solution was used for each experiment, which was used as the electrolyte. The pH of the electrolyte was adjusted, if required, with 1 M HCl or 1 M NaOH (Analar Reagent, Merck, Germany) solutions before experiments. To examine the effect of co-existing ions, for the removal of phenol, sodium salts (Analar Reagent, Merck, Germany) of carbonate, phosphate and silicate were added to the electrolyte for required concentrations.

The analysis of phenol was carried out UV–vis Spectrophotometer (MERCK, Spectroquant Pharo 300, Germany) using Merck test kits. The SEM of mild steel was analyzed with a scanning electron microscope (SEM) made by Hitachi (model s-3000 h).

3. Results and discussion

3.1. Effect of initial concentration of phenol

The effect of concentration of phenol on its removal from the water in the concentration range of 0.5–5 mg/L is presented in Fig. 2. From the figure it is seen that as the phenol concentration increases, the removal efficiency decreases from 98, 94, 92, 76 and

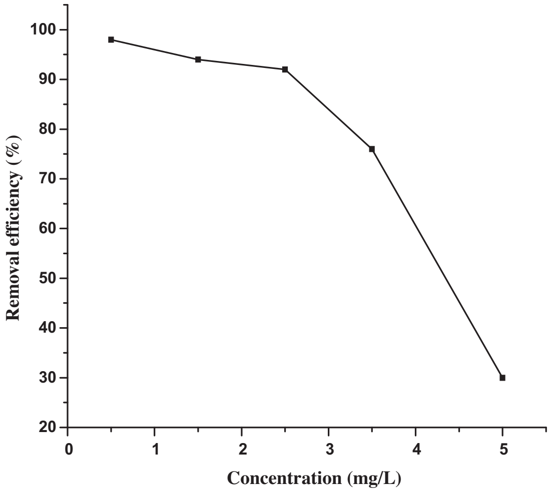

Fig. 2. Effect of initial concentration on the removal efficiency of phenol from water. Conditions, solution pH: 2.0; solution temperature: 303 K; anode: mild steel; cathode: graphite; current density: 0.10 A/dm^2; duration: 30 min.