PHENOL REMOVAL USING ELECTROLYTIC CELL WITH GRAPHITE ANODE

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Abstract: Phenol and phenolic compounds are refractory and toxic. Electrochemical process with inert anode can induce oxidation of organic compounds. This study investigated electrooxidation of phenol in a 600 ml batch reactor. Electrolytic cell comprised of graphite anode and iron cathode at 2 and 6 cm distance. Phenol concentration and COD were analyzed, by colorimetric methods, to determine the effect of NaCl, voltage and reaction time. It was found that in the absence of NaCl, very little phenol was removed. Addition of 1 and 3 gNaCl/l increased the removal to 35% and 56% repectively at the driving voltage of 15 volts. Also, increasing of voltage from 7 to 10 and 15 resulted in increasing removal from 30% to 50% to 56% respectively. Removal of COD was in the range of 4.3-6.8%. Significant difference in removal of phenol and COD indicated that phenol was partially oxidized with oxidation products remaining in other organic forms.

***Keywords**:* phenol; electrooxidation; NaCl; graphite anode

Introduction: Oxidation reactions can be induced by electrochemical process using inert electrodes. Electrochemical process reduces the need for chemical handling and storage. Oxidation can take place either directly or indirectly. In direct oxidation, organic compound is oxidized directly at the anode. In indirect oxidation, electrolyte present in the system is oxidized and converted to secondary oxidant which then oxidizes the organic compounds [1]. Important factors affecting electrooxidation include types of electrode, electrolyte, and electrical density [2]. Various electrode materials had been tried for electrooxidation such as graphite, platinum, titanium oxide, etc. Experiments on electrooxidation of many types of organic pollutants has been carried out. Satisfying results were obtained in the treatment of recalcitrant wastewater such as textile wastewater [3], tannery wastewater [4] and landfill leachate [5].

Phenol and phenolic compound is an organic compound widely used in various industries, such as petroleum refineries, synthetic chemical plants, pulp and paper, textiles, pesticide and herbicide, and pharmaceutical factories. With aromatic structure, phenol is refractory in nature. It is highly toxic. Electrooxidation is an attractive method for the treatment of refractory pollutants, including phenol. Electrode materials were found to play an important role on the oxidation reactions. Electrolytes present in the system also affect the product and extent of oxidation. In the presence of sufficient hydroxyl radicals, phenol and its oxidation products could be completely oxidized to CO_2 [6,7].

The aim of this work was to investigate the performance of graphite-iron electrode electrooxidation process. Graphite was used as anode, iron plate as cathode and NaCl as electrolyte. Phenol concentration in the samples used all through the experiment was 150 mg/l. Variations in the experiment included driving voltage, NaCl concentration and distance between the two electrodes. In addition to phenol determination, Chemical Oxygen Demand (COD) was also determined and used for investigation of organic contents.

Materials and Methods: The experiment was conducted in a batch reactor made of clear acrylic tank. The reactor size was $17x17x25cm^3$ with 600 mL capacity. The anode and cathode plates of 15x20 cm^2 surface area were positioned vertically, parallel to each other. Reactor set up is as shown in Fig 1. Electrodes were connected to voltage adjustable DC power supply.

The sample used in the experiment was phenol solution of 150 mg/l initial concentration, prepared from reagent grade phenol. NaCl was added in to serve as an electrolyte. In each experiment, 600 ml sample with NaCl was transferred into the reactor. A driving voltage was applied. After a pre-set period (at 0, 15, 30 and 60 min.) a sample was drawn out for COD and phenol analysis, using colorimetric method according to APHA standard methods [8]. The parameters used in the study were: NaCl concentration (0, 1 and 3 g/l), electrode distance (2 cm. and 6 cm.) and driving voltage (7, 10 and 15 volts).

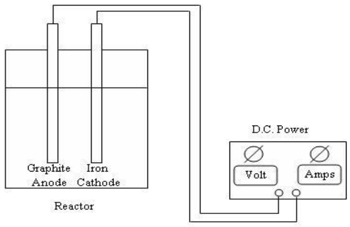
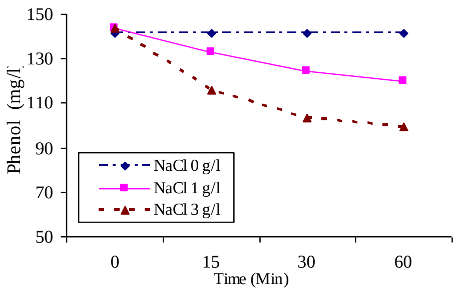


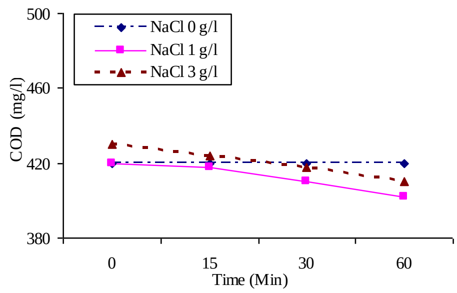
Figure 1. Schematic diagram of experiment set up (electrolytic cell)

Results and Discussion:

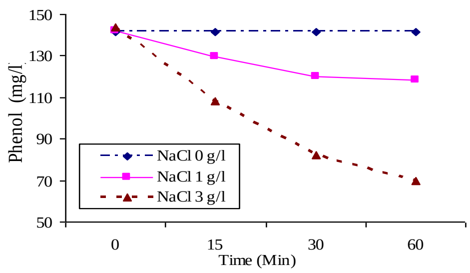
The effect of electrooxidation process on phenol concentration and COD was investigated. The results of the experiments are shown in Fig 2 (a) - (f).



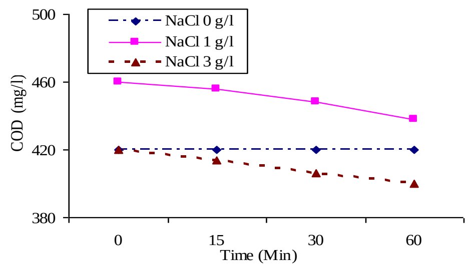
(a) 2 cm, 7volts



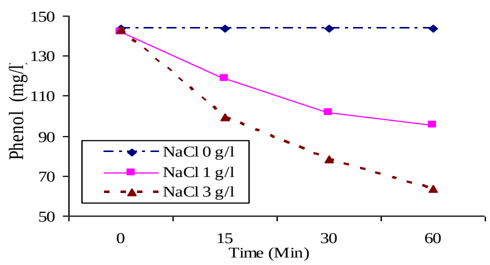
(b) 2 cm, 7volts



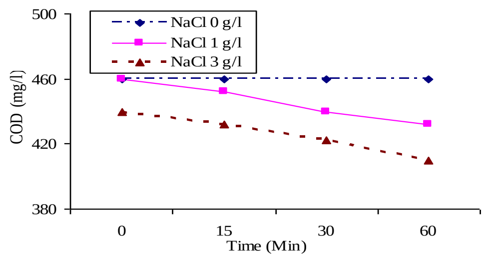
(c) 2 cm, 10volts



(d) 2 cm, 10volts



(e) 2 cm, 15volts



(f) 2 cm, 15volts

Figure 2 . Effect of electrooxidation on phenol and COD at 2 cm. electrode distance

1. The effect of electrooxidation on phenol

The increase in reaction time meant an increase in the charge transfer. For all experiments, phenol concentration decreased with time. The longest period in this study, 60 min., gave the highest removal.

Each graph in Fig. 2 compared treatment results of 0, 1 and 3 gNaCl/l at the same voltage. The results from all 3 voltages followed the same trend. Very little phenol was removed in the absence of NaCl. The removal was enhanced by the addition of NaCl. Phenol remaining at 15 volts, 60 minutes retention time, decreased from 95.6 with 1 gNaCl/l, to 63.6 mgPhenol/l with 3 gNaCl/l. The same trend was observed at 7 and 10 volts.

With regard to the voltage, it can be seen that higher amount of phenol was removed with higher voltage. Phenol remaining at 15 minutes retention time, with 3 gNaCl/l, changed from 116 to 108 to 99 mgPhenol/l when the voltage was changed from 7 to 10 to 15 volts. The same trend was observed at 30 and 60 minutes retention time.

2. The effect of electrooxidation on COD

It can be seen that only a slight amount of COD was reduced over the reaction period. Since COD is the parameter indicating an overall amount of organic matters, not only phenol, the remaining COD indicated that there were still substantial organic matters remaining in the treated samples

3. The effect of electrode distance on phenol removal

The set of experiment with 6 cm electrode distance was also conducted. The same trend was obtained as those of 2 cm. distance. The results at each reaction time are not presented in detail here, but the final results of phenol and COD removal at 60 min of all conditions are compared in Figs.3 and 4.

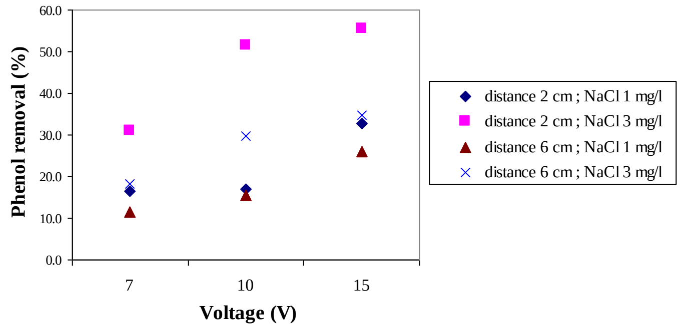


Figure 3. Comparison of phenol removal percentage at 60 min retention time of 2 cm and 6 cm with 1 and 3 gNaCl/l batches

Fig.3 compared the removal percentage of phenol. It can be seen that phenol removal at 2 cm distance was superior to those of the 6 cm. one. At 7, 10, 15 volts with 3 gNaCl/l, 30%, 50% and 56% phenol were removed in the 2 cm batch but only 18%, 30% and 34% in the 6 cm batch.

The COD removal percentage was compared in Fig.4. Since very low COD removal percentages were achieved in all batches, the differences among them were therefore very small. In the 6 cm batch at 15 volts with 3 gNaCl/l, COD could be removed by 4.8 – 5.8% close to 4.6-6.8% in the 2 cm batches.

While a significant amount of phenol was removed, COD was reduced only slightly. The high amount of COD attained over the reaction time showed that other organic compounds were present in the treated sample. This revealed that phenol was not completely oxidized but only changed to other forms of organic matters. Comninellis and Pulgarin [7] reported the detection of maleic acid and oxalic acid as reaction products of electrooxidation of phenol.

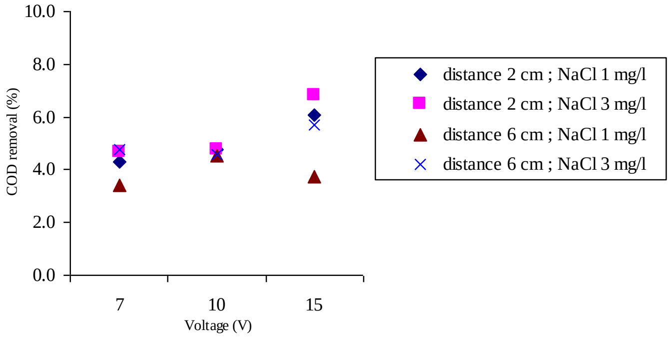


Figure 4. Comparison of COD removal percentage at 60 min retention time of 2 cm and 6 cm with 1 and 3 gNaCl/l batches

CONCLUSIONS : From the experiment, it can be concluded that phenol could be removed by electrooxidation process with graphite anode. The distance between anode and cathode, the driving voltage and NaCl concentration significantly affected phenol removal. At 60 min reaction time, 2 cm electrode distance with 3 gNaCl/l, 56% phenol was removed, while only 36% was achieved in the 6 cm distance batch. In all the treatment conditions tried in the study, very low COD removal of 4.6-6.8% could be achieved. The significant difference of phenol and COD revealed that phenol was not completely oxidized under the conditions studied.

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References:

[1] J. Grimm, D. Bessarabov, and R. Sanderson. 1998. Review of electro assisted methods for water purification. Desalination. 115(3):285-294.
[2] C. Comninellis. 1994. Electrocatalysis in the electrochemical conversion/combustion of organic pollutants for wastewater treatment. Electrochimica Acta. 39(11/12):1857-1865.
[3] J. Naumczyk, L.Szpyrkowicz and F. Zilio-Grandi. 1996. Electrochmical treatment of textile wastewater. Water Science and Technology. 34(17):17-24.
[4] N.N. Rao, K.M. Somasekhar, S.N. Kaul and L. Szpyrkowicz. 2001. Electrochmical oxidation of tannery wastewater. J. of Chem. Tech. and Biotech. 76(11):1124-1131.
[5] L.C. Chiang, J.E. Chang and T.C. Weng. 1995. Electrochemical treatability of refractory pollutants in landfill leachate. Hazardous Waste & Hazardous Materials. 12(1):71-82.
[6] Xiao-yan Li, Yu-hong Cui, Yu-jie Feng, Zhao-ming Xie and Ji-Dong Gu. 2005. Reaction pathways and mechanisms of the electrochemical degradation of phenol on different electrodes. Water Research. 39:1972-1981.
[7] C. Comninellis and C. Pulgarin. 1991. Anodic-oxidation of phenol for wastewater treatment. J. Appl.Electrochem.27: 970-974.
[8] APHA, AWWA and WPCF. 1998. *Standard Methods for the Examination of Water and Wastewater. 20th ed.* APHA.Inc. Wahington D.C.