

## Treatment of Phenolic Effluent by Ozonation: A Review

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### Abstract

The effluent from product processing and refining plants contains phenol and phenolic compounds which are harmful and toxic in nature, requires a great attention in wastewater treatment. Ozone is a strong oxidizing agent, the reaction of it with organic and inorganic ions in aqueous media has achieved a verity of treatment goals. This review paper focuses on the comparison of various advanced oxidation processes for degradation of phenol and phenolic compounds, effects of ozonation, catalytic ozonation and economic aspects of different phenol removal processes.

**Keywords:** Treatment, Phenol, Effluent, Ozonation, Catalyst, Chemical Oxygen Demand

### 1. INTRODUCTION

In the current scenario several acute global issues came up like, global warming, fresh water crisis, environmental pollution, dreadful diseases etc. Environmental pollution is a worldwide problem that is faced by developing and developed countries in recent years. Due to the rapid industrialization increases in generation of waste streams which affects the environment and human health. [1, 11]

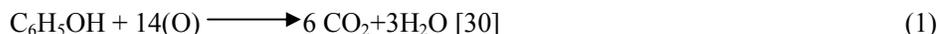
The release of large quantities of polluted industrial wastewater to the environment creates the need for improvement in existing technologies through developing and implementing green, suitable and sustainable treatment technologies that removes hazardous contaminants present in many of these industrial streams. [6, 11, 22]

Phenols, defined as hydroxyl derivatives of benzene. [29] The existence of phenol or phenolic compounds in water can be attributed to natural and anthropogenic activities. Natural sources of phenolic compounds in water pollution include decomposition of organic matter, synthesis by microorganisms and plant in the aquatic environment. Anthropogenic sources of phenolics in water pollution include industrial, domestic, agricultural and municipal waste. [31]

Phenol and phenolic compounds are common organic constituents of many industrial effluents because of it can be used as an intermediate chemical in a wide range of applications. [1] The toxic levels usually range between the concentration of 10-24mg/l for human and 9-25mg/l for fish. Lethal blood concentration of phenol is around 150mg/100ml and because of that effluent containing phenolic compounds cannot simply be released into the environment without treatment. [22] Phenolic compounds can be reduced or removed by various methods like stream gas stripping, adsorption/ion exchange, solvent extraction, membrane based separation method, biological treatment, oxidation processes etc.

USEPA has listed phenol as priority pollutant and set a discharge limit of 0.1 mg/l of phenol in wastewaters.[13] As per Indian Standards for disposal of treated effluents (IS: 2490 Part A), the permissible limit for phenol for the discharge of effluent into inland surface water is 1.0 mg/l and in public sewer and marine disposal it is 5 mg/L.[4]

Due to high oxidation and disinfection potential of ozone, it is widely used for water and wastewater treatment. To improve color and taste of water as well as to remove organic and inorganic compounds from water and wastewater, ozone has been applied. [20] Ozone molecules break down recalcitrant and toxic organic compounds like phenol into smaller molecules.[23]Ozonation process of phenol accomplished through direct ozone oxidation and indirect free hydroxyl radical oxidation. [23]The reaction process has been mentioned below



## 2. Literature Review

### 2.1 Comparison of Various Advanced Oxidation Processes for Degradation of Phenol and Phenolic Compounds

**Santiago Esplugas et. al. (2002)** studied and compared different AOPs (ozone and its combination, photolysis and UV/H<sub>2</sub>O<sub>2</sub>, Fenton and photocatalysis) with a view to the pseudo-first order kinetic constant and cost estimation for the degradation of phenol in aqueous solution. High-energy cost was obtained with photocatalysis and UV based oxidation processes. Based on degradation rate and the costs obtained with ozonation, it appears to be the most attractive option for phenol degradation.[19]

**P. Saritha et. al. (2007)** have studied the advanced oxidation processes (UV, H<sub>2</sub>O<sub>2</sub>, UV/H<sub>2</sub>O<sub>2</sub>, Fenton, UV/Fenton and UV/TiO<sub>2</sub>) for the removal of 4- chloro-2 nitrophenol. The degradation trends followed the order: UV/Fenton > UV/TiO<sub>2</sub> > UV/H<sub>2</sub>O<sub>2</sub> > Fenton > H<sub>2</sub>O<sub>2</sub> > UV. It was concluded that the UV/Fenton was the most effective in partial mineralization of 4C-2-NP. Though, lower costs were obtained with H<sub>2</sub>O<sub>2</sub>. [16]

**Ana Paula Barbosa Rodrigues de Freitas et. al. (2013)** concluded that the process of ozonation for the removal of total phenols was highly efficient thus; it contributes to the increase in effluent biodegradability and the absence of ozone in the reaction medium. [2]

**Haidong Zhou et. al. (2015)** studied the removal of four typical organic micro-pollutants in effluent sewage by individual ozonation (O<sub>3</sub>), ultrasonic ozonation (US/O<sub>3</sub>) and photocatalytic ozonation (PC/O<sub>3</sub>). It was observed that the removal rates of the compounds increased with the rise on pH value in the set range (3.7–9.5) of this study. It was observed that the US/O<sub>3</sub> combined process could achieve the best removal of the target micropollutants. [8]

### 2.2 Effects of Ozonation of Phenol and Phenolics Present in Water or Wastewater

**Anna Goi et. al. (2004)** studied the degradation of nitrophenols (NPs) in terms of biodegradability improvement, and toxicity reduction. The ozone consumption obtained in this study was 1.14 and 1.36 mol of ozone per mol of NP degraded at initial pH 9.5 and 2.5, respectively. COD reduction (38–80%) was lower than NPs removal (77–100%). It was concluded that the initial pH 9.5 was more favourable for the ozonation of the NPs mixture than pH 2.5. The by-products of 4-NP ozonation at pH 2.5 and 2,4-DNP ozonation at initial pH 9.5 were completely biodegradable. After 77-100% conversion of single NPs the degree of nitrogen mineralization was 66-100% at pH 2.5 and 38-87% at initial pH 9.5. It was observed that up to at least 80% degradation of NPs led to the 9.8 (4-NP, initial

pH 9.5), 5.3 (2,4-DNP, initial pH 9.5), and 11.8% (2-methyl-4,6-dinitrophenol, pH2.5) removal of TOC.[3]

**Yung-Chien Hsu et. al. (2004)** investigated the effects of pre-ozonation on the biodegradability of 4-cresol, 4-nitrophenol and 2-chlorophenol solutions using a new gas-inducing reactor with high ozone utilization rate. Combination of preozonation with bio-treatment would provide a feasible means for the treatment of phenolic wastewater. It was concluded that the biodegradability of preozonized phenolic solutions were powerfully dependent on the decomposition of phenolic compounds and accumulation of intermediate products. [25]

**D. Manojlovic et. al. (2007)** studied the removal of phenol and chlorophenols from water by new ozone generator. The ozone generator was based on coaxial dielectric barrier-discharge and operated in the air at an atmospheric pressure. Most of the phenol was removed during ozonation (89.5%) from Sample A (distilled water). After 24 and 96 hours, the percentage of phenol removed was increased to 93.3 and 98.9%, respectively. With 4-chlorophenol and 2,4-dichlorophenol, 99.8 and 98.9% of corresponding phenols were removed. In experiments with two different water samples from Danube (D,E), the phenol was also mostly removed during ozonation, 76.7 and 80.0%, respectively. [5]

**Kadir Turhan and SuheylaUzman (2008)** have investigated that the degradability of phenol in aqueous solutions using ozonation. It was concluded that the system could get free of phenol to part it with ozonation process. Degradation was computable with first degree kinetic. It was seen that some organic compounds such as, catechol, hydroquinone, p-benzoquinone can be oxidized completely with ozone to CO<sub>2</sub> and H<sub>2</sub>O, but destruction of some organic compounds requires a long ozonation time and high ozone dosages. [10]

**Pieter Van Aken et. al. (2015)** investigated the degradation of 2, 4-dichlorophenol (2,4-DCP), as toxic model pollutant in wastewater by ozonation, in the presence of readily biodegradable substrates. After pre-treatment of ozonation, 2, 4-DCP was decreased. As the biodegradable fraction increases and the amount of refractory COD decreases by the ozone treatment because of the (partial) degradation of the pollutant. The toxicity effects of 2,4-DCP decreased due to the ozonation.[18]

**Shibo Wang et. al. (2016)** studied the degradation of phenol in wastewater with ozone produced by self-design ozone generator. The optimized tube's structure of the self-design ozone generator was made with the double dielectric inner electrode and small metal cones were embedded in the outside electrode. When the concentration of phenol in wastewater was 100mg/L, the phenol removed 99.9% in the oxide time for 30 min. The results showed that the phenol removal rate increased as the reaction time increased. As the initial pH increased in wastewater, the phenol removal rate was increased. p-benzoquinone, 4-cyclopentene-1, 3-dione and diacetone alcohol were determined as the major degradation product of phenol. [21]

### 2.3 Catalytic Ozonation of Phenol and Phenolic Compounds

**Nor Aishah Saidina Amin et. al. (2005)** studied the removal of aqueous phenol and COD by ozonation, adsorption and catalytic ozonation with granular activated carbon (GAC) and alumina. It was concluded that presence of ozone enhanced the phenol and COD removal achieved by GAC and alumina. Result showed that both the GAC and alumina gave better results than ozone alone with complete (100%) and 95.1 % phenol removal respectively, compared to 63.1 % removal achieved by ozone. It was observed that the COD removal also increased with the combined system of ozone with either GAC or alumina. The GAC gave higher percentage of COD removal than alumina. [14]

**Yuming Dong et. al. (2008)** studied the catalytic activity and stability of Y zeolite for phenol degradation in the presence of ozone. The flow rate of oxygen was 5 mL/ min, and the flow rate of ozone was 0.30 mg/ min. It was observed that the catalyst was also helpful for the degradation of toxic intermediates. The result indicated that Yzeolite accelerated the decomposition of ozone and the

generation of hydroxyl radicals, consequently enhanced the degradation of phenol and the removal of COD.[24]

**Gholamreza Moussavi et. al. (2009)** investigated the removal of phenol from saline wastewater by catalytic ozonation and integrated ozonation / biological processes. It was found that NaCl had no adverse effect on phenol removal at ranges between 0.5 and 50 g/L. Result showed that COP in the presence of activated carbon was able to degrade almost all the phenol and remove a considerable of COD (80%) from saline wastewater after 60 min reaction time and concluded that long time requirement would impose higher cost on the treatment plant.[7]

**Parvin Gharbani, Ali Mehrizad (2014)** investigated that efficiency of nanosized ZnO in the catalytic ozonation of 4-chloro-2-nitrophenol and determined the effect of pH on heterogeneous catalytic ozonation. Result indicated that after 2 min., the degradation efficiency of 4C2NP was as high as 85.2% at pH3, while the degradation efficiency of 4C2NP was about 31–49% at pH 7-9. It also indicated that the presence of ZnO, accelerated the degradation of 4C2NP rather than the simple ozonation. It was concluded that at the end of the process, degradation of 4C2NP by nano-ZnO (34.9%) was greater than those of the micrometer size (5.8%) due to increase of surface area. In catalytic ozonation, the degraded 4C2NP by nano and micro ZnO was about 99.4% and 91.7%, respectively (at the first 5 min). It was also observed that the concentration of nitrate formed during nano-ZnO catalytic ozonation was 7.08 mg/ L at pH3 and the amount of total organic carbon was 54.9% after 30 min. [17]

**Zichuan Ma et. al. (2014)** investigated the activity of the synthesized catalysts for ozonation of p-nitrophenol (PNP) in aqueous solution. The heterogeneous reaction mechanism involved the adsorption of ozone and organic molecules, and the surface reactions between them. It was concluded that Mn–Co–Fe with proper surface properties exhibited highest catalytic activity and stability for the removal of organic pollutants. [28]

**Zhaokun Xiong et. al. (2016)** investigated the degradation of PNP by  $m\text{Fe}^0/\text{O}_3$  and compared with conventional  $\text{Fe}^0$ , ozonation and  $\text{Fe}^0/\text{air}$  processes. The optimal operational parameters (initial pH of 5.3, ozone flow rate of 1.5 L/min, and  $\text{Fe}^0$  dosage of 40 g/L) of the  $m\text{Fe}^0/\text{O}_3$  process were achieved by the semibatch experiments. The COD removal (89.5%) was obtained by the  $m\text{Fe}^0/\text{O}_3$  process was about twice of the sum (44.8%) of COD removal by  $\text{Fe}^0$  alone and  $\text{O}_3$  alone after one hour. It was observed that COD removal efficiency obtained by the  $m\text{Fe}^0/\text{O}_3$  process (89.5% after 60 min treatment) was also much higher than those of the  $\text{Fe}^0/\text{air}$  process (17.5%),  $\text{MnO}_2/\text{O}_3$  (45.5%) and  $\text{Al}_2\text{O}_3/\text{O}_3$  (74.4%). It was concluded that the high-efficient  $m\text{Fe}^0/\text{O}_3$  process was mainly resulted from the combination of homogeneous ( $\text{Fe}^{+2}$ ,  $\text{Fe}^{+3}$ ) or heterogeneous ( $\text{Fe}_2\text{O}_3$ ,  $\text{Fe}_3\text{O}_4$ ) catalytic ozonation, Fenton-like reaction, adsorption and precipitate. [26]

## 2.4 Treatment of Various Industrial Effluent by Ozonation

**Hoda Roushdy Guendy (2007)** studied the decolorization of two types of dyes, Direct Pink 3B and Reactive Violet SH- 2R, by ozonation in aqueous medium. As the dye concentration increases, the time require for both dyes to decolorize increases. For Reactive dye the lower pH was much better. While in case of Direct Pink the suitable pH was found to be in alkaline medium (7-10). As the current increases, the time of decolorization and amount of ozone decreases sharply for each dye. It was found that as ozone-air flow rate increases the amount of reacted ozone and time of ozonation decreases slightly in both the dyes. Study suggested that the relationship between temperature and stability and solubility of ozone in water was inversely proportional. Increase in temperature reduces the ozone solubility and stability in water. It was found that 98% decolorization occurred in few minutes. [9s]

**Marcia Regina Assalin et. al. (2009)** evaluated the treatment of Kraft effluent by combining in sequence activated sludge and ozone processes. Result showed that the combination of activated

sludge process and ozonation at pH 10 was the most efficient process, due to the lower ozone consumption. It was concluded that the combination of both the processes at pH10 was able to remove more than 80% COD, TOC and color from Kraft E<sub>1</sub> effluent. The efficiency of phenol removal was around 70%. The ozonation post-treatment carried out at pH 8.3 also showed better results than the single process. It was found that the difference in the concentration of free radical produced by activated sludge-O<sub>3</sub>/pH 10 and activated sludge-O<sub>3</sub>/pH 8.3 affected mainly the TOC and total phenol removal values.[12]

**P. Gharbani et. al.(2010)** studied the degradation of 4- chloro-2-nitro phenol by ozonation in aqueous solution in a semi batch reactor under constant ozone dose and variable pH conditions. Ozonation was observed to be more effective in alkaline reaction of medium than other conditions. A significant improvement in chemical oxygen demand removal was observed at pH above 7. At the end of the process, 56.9% COD removal was obtained at pH 9. At pH 3, the degree of organically bounded nitrogen conversion to nitrate was higher. It was concluded that the total organic carbon reduction was 15.89 % at pH 9. The main intermediate product from 4-chloro-2-nitro phenol degradation was chlorophenol. [15]

**Zhenglong Xiong et. al.(2010)** studied the heterocyclic pesticide effluent using ultrasonic/ozone combined process for increasing biodegradability and reducing biological toxicity. It was concluded that the ultrasonic frequency, initial pH of the solution and ozone dosage had a great influence on the COD removal efficiency. The influence of ultrasonic power and ultrasonic probe diameter was small. Low ultrasonic frequency brought better COD removal. It was found that alkaline conditions were more favourable for COD removal. The BOD<sub>5</sub>/COD of the pesticide effluent was increased from 0.03 to 0.55 and the EC<sub>50</sub> was increased from 11% to 52% by the ultrasonic (20 kHz)/O<sub>3</sub> treatment. This results indicated that a considerable improvement in the biodegradability of the effluent and a major reduction in the biological toxicity. [27]

### 3. Concluding Remarks

Ozonation process is being used in industries for TOC, COD, phenol and color removal. pH, ozone dose, temperature, contact time are the major factors affecting parameters for phenol and COD removal by ozonation. In single ozonation process alkaline pH is more favourable, which results in higher phenol and COD removal. In the Catalytic ozonation, process rates increase by decreasing reaction time.

Ozonation is effective for detoxification and biodegradability improvement of wastewater which contains phenolic compounds. The economic feasibility of individual ozonation process is much adoptable compare to other process like catalytic ozonation and other advanced oxidation process. Combination of two or more advanced oxidation processes, enhances free radical generation, which leads to higher oxidation rates but it consumes more energy.

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