

## Case Report

# The Use of E-Peroxone to Neutralize Wastewater from Medical Facilities at a Laboratory Scale

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**Abstract:** The treatment of medical wastewater by the peroxone (AOP) and electro-peroxone (E-peroxone) processes was analyzed. The E-peroxone process is based on the production of hydrogen peroxide electrochemically from an O<sub>2</sub> and O<sub>3</sub> gas mixture produced in sparged ozone generator effluent using graphite-polytetrafluorethylene cathodes. The electrogenerated H<sub>2</sub>O<sub>2</sub> reacts with sparged ozone to produce hydroxyl radicals. All advanced oxidation processes presented in this study effectively removed chemical oxygen demand (COD) by up to 87%. The use of E-peroxone showed 15% better results in COD reduction than conventional peroxone. The research suggests that E-peroxone is more sufficient at removing pollutants in wastewater than peroxone. Hence, E-peroxone was found to be more cost-effective than AOP in this case.

**Keywords:** peroxone; medical facilities' waste waters; COD

## 1. Introduction

Wastewater from medical facilities constitutes a significant part of municipal sewage discharge, because the processes of maintaining hygiene conditions in these facilities causes high water consumption. The high level of suspended substances and chemicals, anionic surfactants and organic substances often exceed the norms regarding the quality of municipal wastewater. The surfactants used for disinfection and washing are the main pollutants in medical wastewater. In recent years, the need to reduce the concentration of chemicals, especially surfactants, in wastewater streams is widely promoted due to more stringent environmental conditions and legal provisions. Surfactants are a group of water-soluble and non-water-soluble detergents. Their task is to remove substances difficult to clean (e.g., oils, blood, inorganic pharmaceuticals). The presence of surfactants in wastewater increases COD and makes the process difficult for quick wastewater treatment—their connection with pollutants is permanent and difficult to break down in the wastewater treatment plant. Excessive concentration of these compounds may also contribute to slowing down the processes of biological wastewater treatment or to partial death of the biological deposit. The most commonly used processes for the purification of this type of wastewater are conventional processes such as coagulation, flotation and chemical oxidation or their combinations [1,2].

The conducted research on ozonation of wastewater in order to degrade recalcitrant contaminants does not give satisfactory results [3,4]. In the conventional ozonation process, ozone decomposes to oxygen immediately after its production due to a very short half-life in the aquatic environment [3,4]. This causes problems in the implementation of the process, such as high cost and the requirement for ozone generation on site. Some of the impurities present in the wastewater are persistent for ozone due to its selectivity in the oxidation of some organic compounds [3–6]. For this reason, ozone is often used in combination with other processes and oxidants, such as electrolysis [5,6] and H<sub>2</sub>O<sub>2</sub> [7,8]. These connections increase the efficiency of pollutant degradation. Although the combination of O<sub>3</sub> processes



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and electrolysis have a large potential for removing contaminants, there are contaminants which in comparison with each of these processes separately, continue to generate problems in the process due to the limited adaptability of ozone generators [9].

The use of  $O_3$  and  $H_2O_2$  combination in the associated peroxone production process (AOP) has a synergistic effect on the removal of pollutants [10–12]. Peroxone is used for cleaning soil, ground water and polluted wastewater with volatile organic compounds, polycyclic aromatic compounds, hydrocarbons, petroleum hydrocarbons, chlorinated solvents and metals, ammunition, diesel oil, methyl tert-butyl ether (MTBE), BTEX (benzene, toluene, ethylbenzene and xylene), trinitrotoluene (TNT) and other soluble components from waste [13].

Turkay et al. [14] in their research describe and highlight the advantages of electro-peroxone processes as a combination of two different systems for hydroxyl radical generation (ozonation and electrolysis). The results showed that the combination of ozone and electrolysis with carbon-based cathodes in optimised conditions will lead to higher degradation, removal rates, and efficiencies compared to using ozonation and electrolysis individually. The comparison tests conducted by Donghai et al. revealed that the coupling of electrolysis and ozonation could synergistically produce hydroxyl radicals ( $HO\bullet$ ) and the separation of cathodic reactions and anodic oxidations further promoted  $HO\bullet$  generation, which was responsible for the enhancement of PABA elimination in the compartmental E-peroxone process [15].

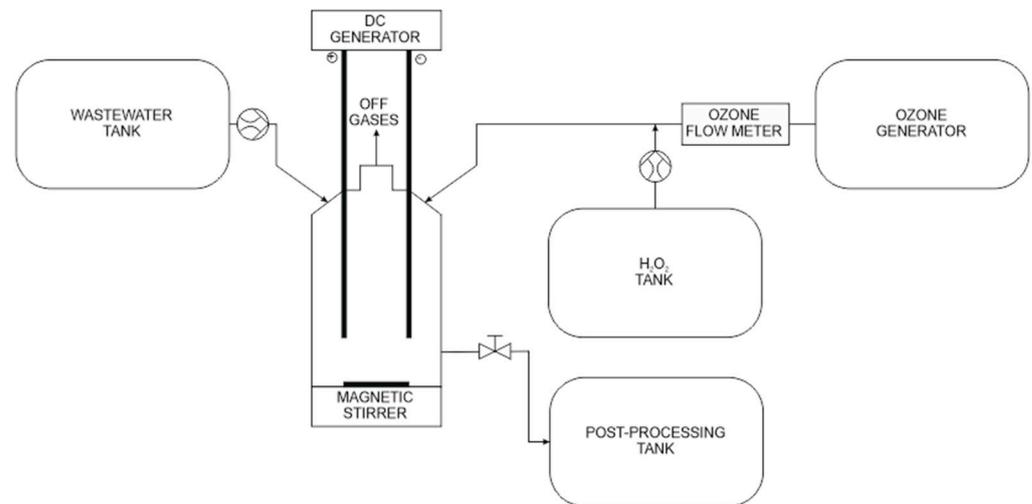
Electro-peroxone (E-peroxone) is an advanced method of electrochemical wastewater oxidation. In this method, the production of  $H_2O_2$  occurs in controlled conditions, in contrast to the conventional oxidation process. The main advantage of using E-peroxone is the use of graphite cathode to form  $H_2O_2$  from water and oxygen in the gas mixture ( $O_3$  and  $O_2$ ). Hence, this process is simple in application, cost-effective and safe. The effectiveness of this process was evaluated in the neutralization of oxalic acid [16], 1,4-dioxane [17], methylene blue [18], Orange II [19] and numerous pharmaceuticals [20]. All studies concentrated mainly on organic substances and results were highly effective. The innovative factor in the conducted research is the use of E-peroxone technology in the process of neutralizing medical wastewater. This issue has not been deeply described in the literature and the potential for using the above-mentioned technology as an alternative to commonly used solutions is noted.

The purpose of the study was to determine the effect of using E-peroxone on the acceleration of the organic matter biodegradation rate in medical wastewater from healthcare facilities. The biodegradation conditions were controlled over time using COD measurements.

## 2. Materials and Methods

The sample of wastewater was collected from IBC containers, which were delivered to a waste disposal company located in Poland. Sample (0.1 m<sup>3</sup> HDPE container) after collection was stored at 5 °C. After 24 h, conditioning sample was divided into 10 subsamples and subjected to an oxidation process. Before the oxidation process, subsamples were not pretreated. pH of the main sample was measured by an Elmetron CP-511 conductometer with an EPS-1 glass electrode for the measurement of pH in the aqueous solution, in accordance with PN-EN ISO 10523:2012. The initial value of pH was 5.2 units.

The wastewater oxidation process was carried out in a glass laboratory reactor with a capacity of 2.5 dm<sup>3</sup>. The reactor, with the help of electronically controlled dosing systems, was simultaneously treated with wastewater as well as ozone and hydrogen peroxide. The reagents were mixed while dispensing with the use of an electromagnetic stirrer with a constant speed of 1000 rpm. In the process of using E-peroxone, graphite electrodes were additionally mounted to the reactor. The electrodes were supplied with DC current of 500, 1000 and 2000 mA (Figure 1).



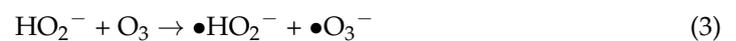
**Figure 1.** Example of experimental set-up.

Each oxidation process lasted 10 min. After each minute of the process, 25 cm<sup>3</sup> of the COD measurement mixture was automatically collected from the reactor discharge port. For each test, the COD index was determined by means of a miniaturized method using sealed tubes according to PN-ISO 15705:2005, repeating each determination three times. Determination of the chemical oxygen demand consists of the introduction of a specified amount of the chemical oxidant and the necessary catalysts and auxiliary substances to the leachate sample. Then, under strict conditions and time, the oxidation process is conducted. After oxidation, the remaining amount of oxidant is determined. The loss of oxidant, converted into oxygen, is given as COD in mgO<sub>2</sub>/dm<sup>3</sup>. The COD was tested in combination with a spectrophotometer (Hach DR-2800). The averaged values of all measurements were assumed for the analysis, after rejecting extreme results (mean value ± 2 times the standard deviation). During testing, the air temperature was maintained at 22 ± 1 °C and air humidity at 55 ± 5%.

Hydrogen peroxide (30%, *w/w*) was of analytical reagent grade (Merck, Darmstadt, Germany). Ozone was produced in the O3PRO30,7VW generator, equipped with a corona electrode system. Gas ozone concentration and flow (in g/Nm<sup>3</sup>) was measured by an ultraviolet gas ozone analyzer, Eltech 200. Ozone in the off-gas was measured by Lenntech AQL S200 analyzer.

### 3. Results

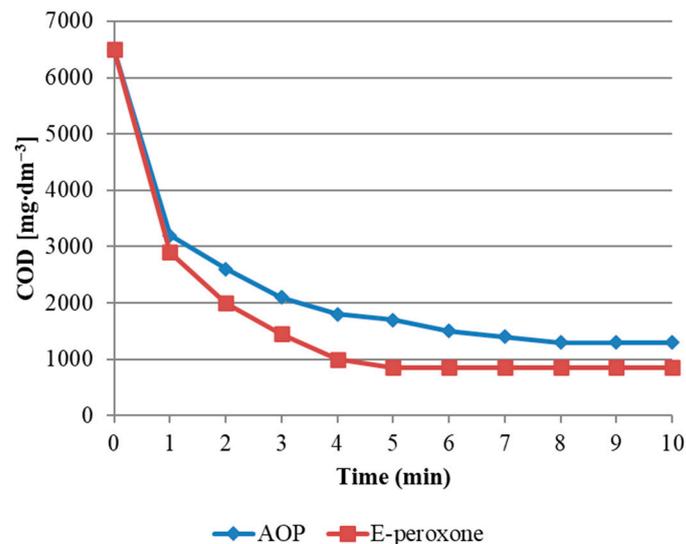
Electro-generation of hydrogen peroxide occurs during ozone presence in the reactor along with wastewater. E-peroxone transforms O<sub>2</sub> gas from O<sub>3</sub> decomposition at graphite cathodes to electro-generate H<sub>2</sub>O<sub>2</sub> (Equation (1)) efficiently. Its conjugated base (Equation (4)) may then react with sparged O<sub>3</sub> gas to form hydroxyl radicals and other radical species (Equations (3)–(8)) [14].





It has been established that the dissociation of  $\text{H}_2\text{O}_2$  (Equation (4)) increases with increasing pH, as the pKa value of  $\text{H}_2\text{O}_2$  was 11.6 [21] and  $\text{H}_2\text{O}_2$  reacts with  $\text{O}_3$  to produce  $\bullet\text{OH}$  only when present as its conjugated base,  $\bullet\text{HO}_2^-$ .

COD reduction in the analyzed wastewater is presented in Figure 2. For the analyzed oxidation processes, a 30% COD reduction between 2 and 3 min of reaction was obtained. After this period, the AOP process slowed down its speed, and E-peroxone proceeded at a similar speed. After completion of the reaction at 10 min, both processes obtained similar COD reduction results, which differed by  $450 \text{ mg}/\text{dm}^3$  COD.



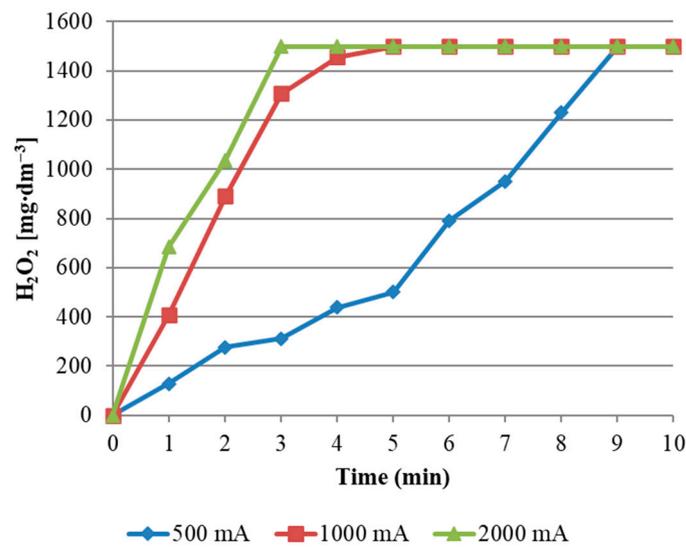
**Figure 2.** COD removal during AOP and E-peroxone processes (experimental conditions: reactor volume of  $2.5 \text{ dm}^3$ ; applied current of  $500 \text{ mA}$ ; ozone flow rate of  $30 \text{ dm}^3 \cdot \text{h}^{-1}$ ; outlet  $\text{O}_3$  gas concentration of  $0.3 \text{ mg}/\text{dm}^3$ ).

According to Figure 2, the regulation of the current in the E-peroxone process resulted in a slight improvement in the kinetics of the oxidation reaction. Studies have shown that the use of a current of  $500$ ,  $1000$  and  $2000 \text{ mA}$  accelerate the COD removal process.

The analysis of the obtained results suggests that the optimal rate of  $\text{H}_2\text{O}_2$  formation occurs when using a current of  $2000 \text{ mA}$ . The use of a higher current intensity applied to the graphite cathode provides faster transformation of the ozone into the hydroxyl radicals in the aqueous environment.

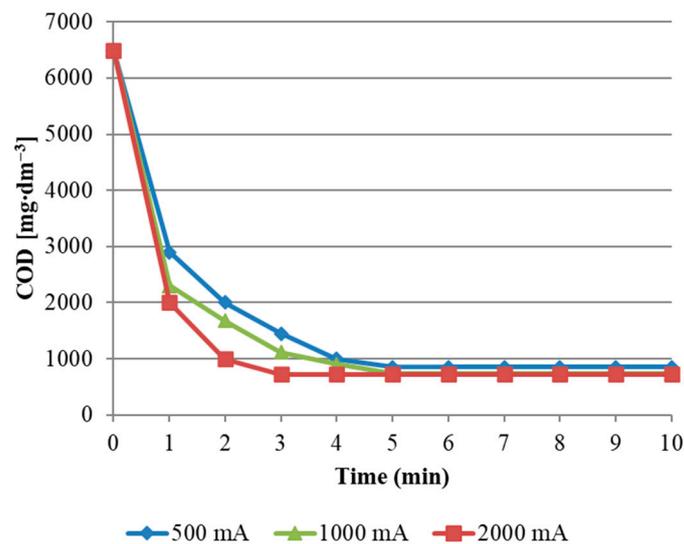
As a result, it is possible to avoid a situation in which the presence of hydrogen peroxide lowers the efficiency of the process, due to the better affinity for hydroxyl radicals than the impurities present in wastewater. This situation is often observed in the AOP process [10,11,22–24].

The influence of current intensity on  $\text{H}_2\text{O}_2$  production in the E-peroxone process is presented in Figure 3. A rapid increase in  $\text{H}_2\text{O}_2$  concentration was observed between the beginning of the process and the 5th minute of its duration. At that time, the concentration of hydrogen peroxide was reached at the level of about  $1500 \text{ mg}/\text{dm}^3$ . This value was the maximum concentration for the current intensity in the range of  $1000$  and  $2000 \text{ mA}$ . The increase in  $\text{H}_2\text{O}_2$  concentration was slower when the applied current was reduced to  $500 \text{ mA}$ . The concentration of hydrogen peroxide generated in the reactor during the use of graphite electrodes reached the equilibrium concentration ( $\sim 1500 \text{ mg}/\text{dm}^3$ ) after  $9 \text{ min}$  of the process for all applied values of current intensity.



**Figure 3.** Electro-generation of H<sub>2</sub>O<sub>2</sub> (experimental conditions: reactor volume of 2.5 dm<sup>3</sup>; applied current of 500, 1000 and 2000 mA; ozone flow rate of 30 dm<sup>3</sup>·h<sup>-1</sup>).

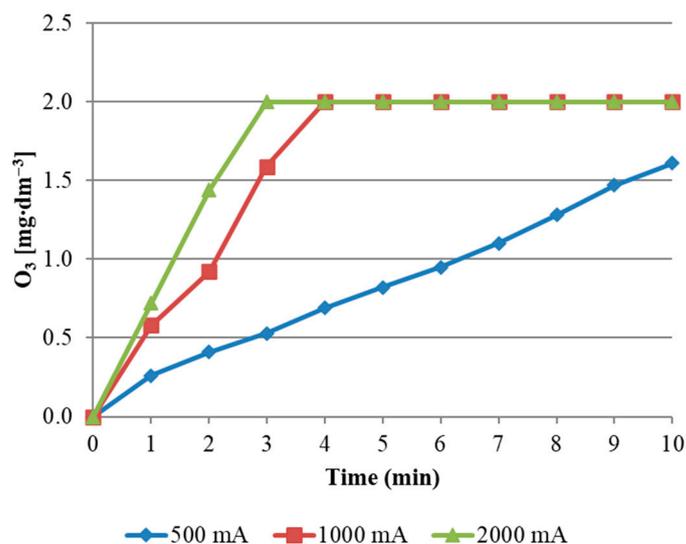
The highest ozone consumption occurred between the beginning of the reaction and the 4th minute of its duration. After this time, the concentration of ozone in the reaction mixture was reached at the level of about 2 mg/dm<sup>3</sup>. This value was the maximum concentration for the current intensity in the range of 1000 and 2000 mA. The increase in O<sub>3</sub> concentration was slower when a current of 500 mA was applied. Comparing the effect of the applied current intensity (Figure 4), it was found that its two-fold increase accelerates the COD reduction.



**Figure 4.** COD removal during E-peroxone processes (experimental conditions: reactor volume of 2.5 dm<sup>3</sup>; applied current of 500, 1000 and 2000 mA; ozone flow rate of 30 dm<sup>3</sup>·h<sup>-1</sup>; hydrogen peroxide start dose 50 cm<sup>3</sup>).

The conducted experiments also show that after reaching the chemical equilibrium in the reactor (5th minute of the process), the current intensity's influence on the course of the reaction is balanced. This observation suggests that the optimal current intensity for wastewater treatment is 1000 mA. Comparing the above statements with Figure 3, it was found that the H<sub>2</sub>O<sub>2</sub> formation rate increases with the current applied to the graphite cathode. As a result, faster and more efficient transformation of ozone into hydroxyl radicals in the E-peroxone system is achieved.

During the tests, it was also found that in the 5th minute of the process hydrogen peroxide begins to take over the action of hydroxyl radicals. This phenomenon confirms the stabilization of  $\text{H}_2\text{O}_2$  concentration and of the content of ozone dissolved in water (Figure 5). An analogous phenomenon occurs when using the standard AOP process described in [11]. As can be seen in Figure 5, the applied current intensity below 1000 mA can provide very different process effects, whereas above this value the effects of the oxidation process are not so diverse. The critical current level may depend on many factors, such as the reaction conditions ( $\text{O}_3$  dose, pH, suspension, etc.) and the properties of the organic impurities present in the wastewater to be neutralized.



**Figure 5.** The change of aqueous  $\text{O}_3$  concentration during electro-generation of  $\text{H}_2\text{O}_2$  (experimental conditions: reactor volume of  $2.5 \text{ dm}^3$ ; applied current of 500, 1000 and 2000 mA; ozone flow rate of  $30 \text{ dm}^3\cdot\text{h}^{-1}$ ).

#### 4. Summary

The article presents the results of research on the wastewater treatment process from a medical facility, using the conventional and electric peroxone systems. The formation of peroxone with the use of electric current is based on the production of hydrogen peroxide on the graphite cathode. The  $\text{H}_2\text{O}_2$  generated during the reaction reacts with the ozone dissolved in water, causing the formation of highly reactive hydroxyl radicals. In the E-peroxone process the concentration of  $\text{H}_2\text{O}_2$  was maintained at  $1500 \text{ mg}/\text{dm}^3$ , with the flow of ozone through the reactor at the amount of  $30 \text{ dm}^3\cdot\text{h}^{-1}$ .

The use of both processes for wastewater treatment has produced positive results. The use of E-peroxone showed 15% better results in COD reduction than conventional peroxone. For analysis of the cost-intensity of the process, the energy consumption for ozone production and reactor functioning was assumed to be  $5 \text{ kWh}/\text{kgCOD}$ ; cost of electricity to power  $0.25 \text{ EUR}/\text{kWh}$ ; cost of purchasing electrodes,  $10 \text{ EUR}/2\text{pcs}$ ; cost of purchasing hydrogen peroxide with a concentration of 35%,  $400 \text{ EUR}/\text{m}^3$ ; reaction time is 5 min. Based on the above assumptions, the operating cost of the oxidation process using peroxone is on average  $2.54 \text{ EUR}/\text{kgCOD}$ , and  $2.62 \text{ EUR}/\text{kgCOD}$  for the E-peroxone process.

The conducted research confirms the legitimacy of modifying standard oxidation processes that are sufficient to remove most organic pollutants. In practice, before using E-peroxone, it is necessary to consider what kind of wastewater is subject to purification, because its properties will affect the amount of  $\text{H}_2\text{O}_2$  that should be generated during neutralization. Considering that the removal of pollutants occurs most intensively in the first minutes of the oxidation process, it is very important to evenly discharge the leachates and discharge the neutralizer from the reactor. It is not recommended to use long reaction

times, because excessive concentration of peroxone does not significantly affect the quality of the neutralizer.

E-peroxone was found to be an efficient process for removing wastewater pollutants, the efficiency of the process depending on the type and constituents of the wastewater. Besides, an insufficient amount of  $O_3$  and excess amount of  $H_2O_2$  in the solution may reduce the process efficiency. Therefore, the amount of electro-generated  $H_2O_2$  and aqueous  $O_3$  should be controlled during the process.

The presented study is a preliminary experiment provided at the laboratory scale. Obtained results of E-peroxone usage for medical facilities' wastewater will be used at a semi-technological scale to explore the real problems of the proposed process along with economic factors.

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