

# Combined Treatment of Old Sanitary Landfill Leachate

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## Abstract

Landfill leachate presents hardly treatable, highly complex and very toxic environmental effluent originated in the municipal solid waste degradation process. Although, numerous treatment methods were developed so far, none of them alone could achieve permissible limits of the primary pollutants to discharge into natural recipients. The current study aimed to develop and apply the process to treat landfill leachate by simultaneous application of electrochemical methods, ultrasound, electromagnetic field and ozonation to achieve the legal criteria for its discharge into natural recipient and minimize its adverse environmental impacts. For this purpose, old landfill leachate was taken from the Piskornica (Koprivnica, Croatia) sanitary landfill. Prior to the treatment, the leachate was supplemented with NaCl (2 g/L) and subjected to simultaneous treatment with stainless steel electrode plates, ultrasound and recirculation through electromagnetic field. After 45 minutes, stainless steel electrode plates were replaced by iron electrodes and treated for another 10 minutes followed by 15 minutes of the treatment with aluminum electrode plates. Ultrasound and recirculation through electromagnetic field were also applied during Fe and Al electrode treatment. Finally, the electrodes were removed and the suspension was mixed with ozone for another 30 minutes and allowed to settle for an hour. Following the combined treatment, the removal efficiency for the turbidity, color, suspended solids, ammonium, phosphates and heavy metals was 99% or higher, while the removal of COD was 97%. All the measured parameters in the treated leachate were lower compared to upper permissible limit for discharge into natural recipient.

**Keywords:** Electrocoagulation, Electromagnetic Treatment, Electrooxidation, Electroreduction, Landfill Leachate, Ultrasound

## 1. Introduction

The rapid industrial and commercial growth in recent decades in many countries has resulted in the increased amount of both municipal and industrial solid waste (1). The daily waste production per capita usually ranges from 0.5 to 1.0 kg. The largest amount of waste is produced in the developed countries and the countries with the intensive industrial growth. In most countries the common practice of the municipal waste management is disposal onto sanitary landfill. The landfilling of the waste especially onto unregulated disposal sites could cause a significant negative impact on all aspects of the environment, and the most significant risks to the environment are the breakdown products of the waste-landfill gases and landfill leachate (2).

Landfill leachate forms by the complex physicochemical and biochemical transformations of the solid waste deposited at the sanitary landfill as well as the percolation of atmospheric water thorough the waste body of the sanitary landfill (2-5). The volume and the composition of the

leachate depend primarily on the type and the composition of the waste, the age of the landfill, as well as climate parameters. The age of the landfill is one of the key factors affecting the composition of the leachate (3-7). The leachate characteristics are dark color, unpleasant odor, high conductivity, high concentration of organic biorefractory compounds and ammonium as well as increased concentration of heavy metals (2-7).

Numerous treatment methods are developed so far to deoxidise landfill leachate. Among them biological methods based on either aerobic or anaerobic processes, physicochemical methods, membrane technologies, advanced oxidation processes and electrochemical methods are most commonly used (2, 7).

In the last 20 years electrochemical methods are increasingly used to treat wastewaters of low biodegradability and high organic load such as landfill leachate (2). Among electrochemical methods electrochemical oxidation (8-13) is most commonly used to treat landfill leachate due to the possibility of direct oxidation of the heavily degradable organic matter into CO<sub>2</sub> and water. Electro-

oxidation of the organic pollutants is achieved by either direct or indirect anodic oxidation (2). Direct anodic oxidation considers either electrochemical conversion in which electrochemically generated active oxygen is chemically bound in the structure of the anode and is responsible for the chemical conversion of the pollutants while in the case of electrochemical combustion an active oxygen is adsorbed on the surface of the anode in the form of hydroxyl radicals promoting electrochemical combustion. The electrochemical conversion results in partial oxidation of organic matter into more easily degradable compounds suitable for biological treatment while the electrochemical combustion leads to complete destruction of the organic matter into CO<sub>2</sub> and water (2, 3, 5). In the case of indirect anodic oxidation, organic pollutants and ammonia are oxidized with electrochemically generated reactive oxygen species such as chlorine and hypochlorite, hydrogen peroxide and ozone. In case of using iron anode, indirect anodic oxidation of organic pollutants takes place by hydroxyl radicals generated electrochemically in the electro-Fenton reaction from Fe<sup>2+</sup> and hydrogen peroxide formed in situ in electrochemical reactor (2, 5).

In the real conditions the removal of the organic matter and ammonia in the leachate is mostly achieved through the indirect anodic oxidation by free chlorine and sodium hypochlorite generated by the oxidation of chloride (initially present in the wastewater) on the anode. The percentage of the removal depends on the composition of the effluent (depending on the age of the landfill), anode material, current density, concentration of electrolytes (chlorides, sulfates) and the treatment time (2-6).

Among electrochemical methods, electrocoagulation is also successfully used to treat landfill leachate (3, 5, 6, 14-16). Electrocoagulation is highly efficient in the removal of high molecular weight humic substances and suspended solids manifested through significant removal of color and turbidity (3, 5, 6). Depending on the type of electrode, high removal rate is also achieved for heavy metals, phosphates and fluorides.

Considering the complex and variable composition of the leachate, its low biodegradability as well as its high toxicity to various test systems (17-19), a combined treatment approach should be applied to remove toxic pollutants down to the regulated values. In that case, electrochemical methods are combined with advanced oxidation (5) or chemical pretreatment (6) that result in higher removal rates of both COD and ammonium.

The current study aimed to simultaneously apply electrochemical methods, ultrasound and electromagnetic field together with final ozonation to treat landfill leachate to achieve higher removal rate of the organic pollutants and ammonium, and significantly decrease the treatment

time. For this purpose, landfill leachate from old sanitary landfill called Piskornica (Koprivnicki Ivanec, Croatia) was used. The landfill site and its history were described in details in the authors' previous papers (2-6).

## 2. Materials and Methods

### 2.1. Sampling and Sample Handling

For the purification experiments, landfill leachate was collected from the first lagoon of the Piskornica sanitary landfill, Koprivnica, Croatia, in September 2013. The leachate was collected in five polyethylene containers with total volume of 100 L and transported to the laboratory. In order to obtain homogeneous sample the effluent was combined into a single tank and mixed for 10 minutes (600 rpm) before analysis or each purification experiment.

### 2.2. Treatment Experiments

The laboratory scale electrocoagulation (EC) experiments were conducted in quadratic polyethylene vessels. For each experiment, 10 L of the wastewater was taken, supplemented with 2 g/L of NaCl and subjected to simultaneous electrochemical treatment, circulation through electromagnetic field and ultrasound (piezoelectric unit). Electrochemical parameters (current intensity ranging from 20 - 100 A, number of electrodes ranging from 5 - 10, the distance between the electrodes ranging from 5 - 20 mm, mass of supporting electrolyte ranging from 0 - 3 g) for all three electrodes had been optimized in the authors' previous study (3). The leachate was first subjected to electrooxidation/electrocoagulation by stainless steel electrode set (I = 70 A; U = 12 V; reaction time 45 minutes), followed by electroreduction/electrocoagulation using the set of iron (Fe) electrodes (I = 70 A; U = 12 V; reaction time 10 minutes) and finally by electrocoagulation using aluminum (Al) electrode set (I = 50A; U = 12 V; reaction time 15 minutes). Each electrode set contained ten, quadratic, one millimeter thick plates (75 × 250 mm) in the parallel arrangement, separated by electro insulator. The distance between the plates was 10 mm. Each even plate was connected to + pole of DC power supply and represented sacrificial anode, while odd plates were connected to pole of DC served as cathodes. Both sides of each electrode plate represented an active area. During the whole electrochemical treatment, the leachate was circulated through an electromagnetic field and ultrasound unit (42 kHz; 20 W). During the SS treatment the samples were taken every 15 minutes and checked for ammonium concentration. The treatment was stopped when the removal efficiency was less than 5% compared to the previous treatment interval. In the case of Fe

electrodes, the treatment was stopped after complete reduction of  $\text{Cr}^{6+}$  into  $\text{Cr}^{3+}$  assessed by measuring  $\text{Cr}^{6+}$  concentration every two minutes. In the case of Al electrodes, the treatment was stopped when the removal efficiencies of color and turbidity were less than 2% compared to the previous treatment interval and total iron concentration was  $< 0.02$  mg/L. Following the treatment by aluminum electrode set, the suspension was subjected to slow mixing with ozone bubbles supplied by ozone generator model OzoneMax 1668 (500 mg/hour) for 15 minutes to complete coagulation/flocculation, while additional 30 minutes was needed for flock's settlement. All experiments were conducted in triplicate. In all cases RSD was less than 10%.

### 2.3. Chemical Analysis

The parameters such as colour, turbidity, suspended solids (SS),  $\text{NH}_4^+$ , fluorides, phosphates,  $\text{Cr}^{6+}$ , Fe and chemical oxygen demand (COD) were determined by HACH DR890 colorimeter (HACH Company, Loveland, CO). For sample digestion, DRB 200 reactor (HACH Company) was used. Biochemical oxygen demand ( $\text{BOD}_5$ ) (means the test has been run for 5 days) was determined by OxiTop system (WTW, Weilheim, Germany); pH value, electrical conductivity (EC) and total dissolved solids (TDS) were determined by PHT-027 - water quality multiparameter monitor (Kelilong Electron Co Ltd., Fuan Fujian, China) (4).

## 3. Results and Discussion

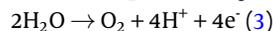
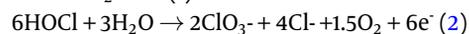
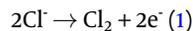
The values of physicochemical parameters in the landfill leachate from the Piskornica landfill and upper permissible limits are presented in Table 1. It was stable leachate with slightly alkaline reaction and dark brown color caused by the presence of high molecular weight humic substances. It was characterized by relatively low COD and  $\text{BOD}_5$  values and low biodegradability ( $\text{BOD}_5/\text{COD} = 0.09$ ). Compared to the regulated values (Table 1) the parameters color, turbidity, suspended solids, COD,  $\text{BOD}_5$ , ammonium and phosphates exceeded the upper permissible limit.

The highest removal efficiencies of all contaminants (with the exception of fluorides and phosphates) were achieved in the first treatment step. After 45 minutes of the combined treatment with stainless steel electrode, ultrasound and electromagnet 90.17% of color, 93.71% of turbidity, 93.74% of suspended solids, 93.10% of COD, 86.76% of  $\text{BOD}_5$ , 98.84% of ammonium and 45.40% phosphate were removed from the leachate (Figure 1). After removing high molecular weight humic substances, the color of the leachate changed from dark brown to yellow.

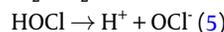
The predominant mechanism of the organic pollutants and ammonium removal was indirect anodic

oxidation with electrochemically generated chlorine and hypochlorite during electrochemical treatment with stainless steel electrode according to the following set of reactions:

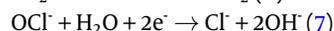
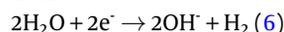
Anode:



Summary reaction:



Cathode:

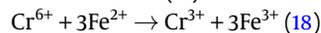
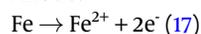


Additionally, sonication of the leachate resulted in rapid formation, growth and collapse of the cavitation bubbles characterized by high temperature (3000 - 5000 K) and pressure (500 - 10,000 atm) leading to either pyrolytic degradation of organic contaminants and ammonium inside the bubbles or their oxidation with free radical species (mostly  $\bullet\text{OH}$ ,  $\bullet\text{OOH}$ ) formed during the cavitation process through thermal dissociation of water and oxygen.

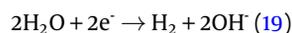
Recirculation through an electromagnetic field resulted in the formation of larger flocs and faster removal of high molecular weight substances (6).

During the treatment with stainless steel electrodes together with  $\text{Fe}^{2+}$  ion, Mn species,  $\text{Cr}^{6+}$  and  $\text{Ni}^{2+}$  ions were also released into solution. Since,  $\text{Cr}^{6+}$  is hardly removable from the solution due to its high solubility, the reduction into less soluble  $\text{Cr}^{3+}$  species was needed for successful removal. This was done by iron electrode plates. The mechanism of  $\text{Cr}^{6+}$  removal could be explained by the potential  $\text{Fe}^{2+}$  ions introduced into solution by electrochemical corrosion of the sacrificial Fe anode plates to reduce  $\text{Cr}^{6+}$  into  $\text{Cr}^{3+}$ . Once converted to  $\text{Cr}^{3+}$  it precipitates as poorly soluble  $\text{Cr}(\text{OH})_3$ . This could be demonstrated by the following reactions:

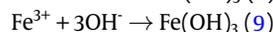
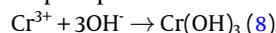
Anode:



Cathode:



Co-precipitation:

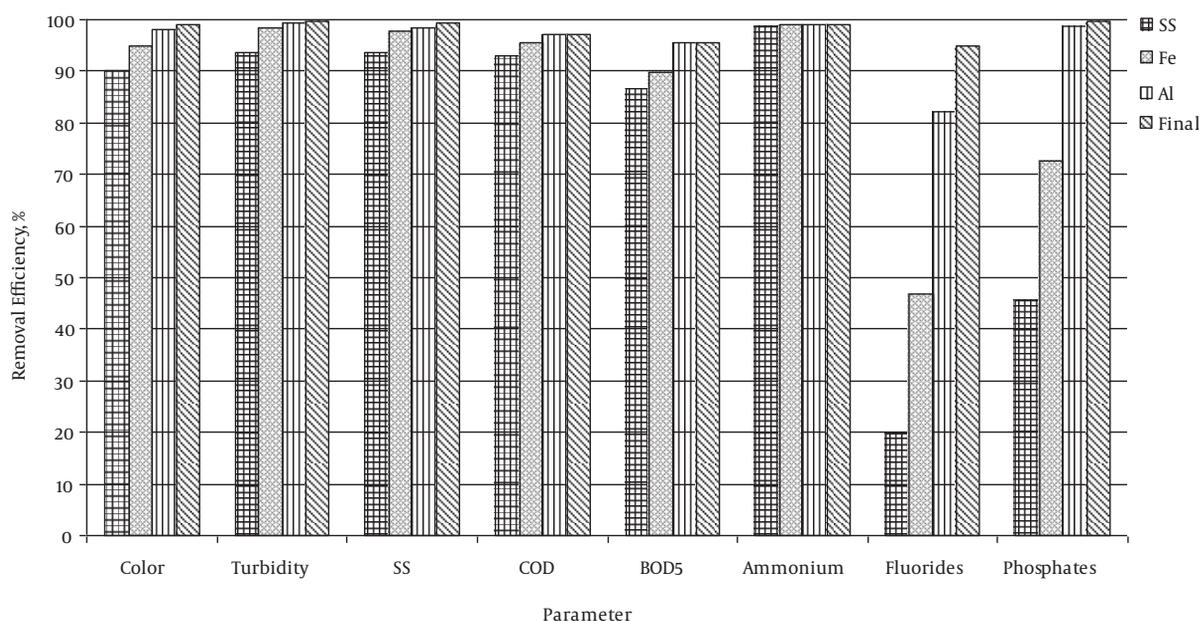


$\text{Fe}(\text{OH})_3$  and  $\text{Fe}(\text{OH})_2$  also serve as the co-precipitant to remove the contaminants (especially heavy metals). Additionally, the bubbles of hydrogen generated by water reduction on the cathode, on its way to the surface, provide constant contact of the flocs with the pollutants in

**Table 1.** Physical and Chemical Parameters Determined in Untreated Landfill Leachate and After Simultaneous Electrochemical, Electromagnetic, Ultrasound Treatment and Ozonation, Together With the UPL for Discharge Into Natural Recipient and Removal Efficiencies for Each Parameter

Parameter	Initial Effluent	Final Effluent	UPL	Removal Efficiency (%)
Color (PtCo)	5400	73	Without	99.02
Turbidity (NTU)	1400	6	Without	99.57
SS, mg/L	591	4	35	99.32
pH	8.21	8.13	6.5-9	-
COD, mg/L	681	19	125	97.21
BOD <sub>5</sub> , mg/L	68	3	25	95.59
NH <sup>4+</sup> , mg/L	129	1.1	10	99.15
Cl <sup>-</sup> , mg/L	794	2730	-	-
F <sup>-</sup> , mg/L	0.391	0.02	10	94.88
PO <sub>4</sub> <sup>3-</sup> , mg/L	3.37	0.01	2	99.70

Abbreviations: BOD, biochemical oxygen demand; COD, chemical oxygen demand; NTU, nephelometric turbidity units; PtCo, the Platinum-Cobalt scale; UPL, upper permissible limit;



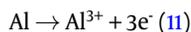
**Figure 1.** The removal efficiencies of the parameters from the landfill leachate after each treatment step. (SS, electrochemical treatment with stainless steel electrode set, ultrasound and electromagnet; Fe, electrochemical treatment with iron electrode set, ultrasound and electromagnet; Al, electrochemical treatment with aluminum electrode set, ultrasound, electromagnet and ozone; final, coagulation/flocculation with electrochemically generated cations and slow mixing with ozone bubbles).

the water, resulting in higher removal rate of the contaminants. Sonication was responsible for the increased removal percentage of organic contaminants and ammonium. Following the treatment with iron electrode plates the removal efficiencies for the parameters color, turbidity, SS, COD, BOD<sub>5</sub>, ammonium, fluorides and phosphates were 94.94%, 98.29%, 97.63%, 95.45%, 89.71%, 99.15%, 46.80% and 72.70%, respectively. Apart from slightly yellow col-

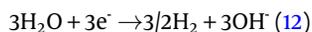
oration originated from the low molecular weight organic substances and excess of Fe<sup>2+</sup>, all other parameters in the leachate were lower compared to the regulated values. To remove those substances aluminum based electrocoagulation was applied for an additional 15 minutes. This treatment step had the highest influence on the removal of fluorides and phosphates. Their removal efficiencies increased to 82.10% and 98.81%, respectively while the removal of

color increased app. 3% compared that of the previous step. The aluminum based electrocoagulation could be explained by the following set of reactions:

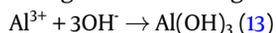
Anode:



Cathode:



$\text{Al}^{3+}$  generated by electrolytic oxidation of the sacrificial anode, and  $\text{OH}^-$  ions generated by the reduction of water at the cathode surface form aluminum hydroxide flocs according to the following reaction:



$\text{Al}(\text{OH})_3$  serves as an adsorbent/co-precipitant for the pollutants in the treated water.

Ozonation of the water resulted in complete removal of odor and oxidation of soluble  $\text{Fe}^{2+}$  into  $\text{Fe}^{3+}$  which was easily removed by coagulation-flocculation resulted in the final concentration of iron < 0.02 mg/L.

The simultaneous treatment using electrochemical methods, sonication, electromagnetic field and final ozonation resulted in clear, colorless and odorless effluent with the concentrations of all parameters of interest below maximum permissible limit for discharge into natural recipient. After 70 minutes of the treatment the removal of color, turbidity, suspended solids, COD,  $\text{BOD}_5$ , ammonium, fluorides and phosphates were 99.02%, 99.57%, 99.32%, 97.21%, 95.59%, 99.15%, 94.88% and 99.70%, respectively.

Direct comparison of the current study results with those of similar studies is impossible due to differences in the treatment methodologies, type of the leachate and initial concentrations of the pollutants. Therefore, only removal efficiencies of the most important parameters (COD and ammonium) could be compared.

Three previous studies (3, 5, 6) were the closest to the present research, since they were conducted on the same leachate using the same electrochemical methods. Electrochemical treatment of landfill leachate (3) using stainless steel, iron and aluminum electrodes with the same setup of the current study, 2 g/L NaCl as the supporting electrolyte, resulted in the removal of 99.31% of color, 99.23% of turbidity, 98.71% of suspended solids, 88.32% of ammonium and 91.97% of COD after 10 hours of the treatment. Another study (5) combined the advanced oxidation, electrochemical and microwave treatment that totally lasted eight hours resulted in the removal efficiency of color, turbidity, suspended solids, ammonium, COD and iron for 98.43%, 99.48%, 98.96%, 98.80%, 94.17% and 98.56%, respectively. By applying pretreatment with CaO and final electrochemical treatment using stainless steel, iron and aluminum electrode plates 94.2% of COD, 99.7%, of color, 99.3% of turbidity, 99.3% of suspended solids and 99.7% of ammonium was removed from the Piskornica leachate after 3.5

hours of treatment (6).

Although similar removal efficiencies were obtained in the current study compared to the previous ones, in the current research the treatment time significantly reduced for the same volume and very similar composition of the leachate.

Compared to the current study results, somewhat lower removal efficiencies of COD (60% to 92%) were obtained by other researchers using electrooxidation methods (8-13). By applying electrooxidation process, 92% of the COD and 100% of ammonium was removed from the old landfill leachate using titanium anode coated with Sn-Pd-Ru oxides (8). The electrooxidation using titanium anode coated with lead oxide (9) resulted in 91.7% of COD, 100% of ammonium and color removal from the leachate. By combining the electrochemical methods 30% - 60% of COD and 70% color was removed from the leachate (10). Approximately 80% of COD was removed from the landfill leachate with initial concentration of 718 mg/L after three hours of the treatment by electro-oxidation (11) using titanium grid coated with lead dioxide layer as anode and stainless steel as cathode. Electrochemical treatment using graphite carbon electrode and  $\text{Na}_2\text{SO}_4$  as an electrolyte resulted in 68% of COD removal from the landfill leachate with initial COD concentration of 1414 mg/L (12). By applying anodic oxidation 69% of COD and 80% of ammonium was removed from the landfill leachate (13).

Even lower removal rate of COD was obtained by electrocoagulation. By applying two types of the electrolytic cells (Al-Cu and Fe-Cu) 30% - 50% of the COD was removed from the landfill leachate with initial COD value ranging from 1134 to 4979 mg/L (14). Using Fe and Al anodes, 87% and 77% of the COD, were removed respectively from the landfill leachate with initial COD value of 4022.5 mg/L (15). After 30 minutes of electrocoagulation by aluminum electrode plates, 45% of COD and 60% of color was removed from the nanofiltration concentrate by-product of the landfill leachate treatment (16).

The removal efficiencies of COD and ammonium reaching 97.21% and 99.15%, respectively obtained in the current study were significantly better compared to those of the previously published data. It was also superior regarding the treatment time.

#### 4. Conclusion

Based on the obtained results it could be concluded that the electrochemical methods combined with the ultrasound and electromagnetic treatment represent suitable treatment approach for the effluents containing complex mixture of the contaminants, especially those with significant amount of recalcitrant organic matter. The

highest removal efficiencies for organic contaminants, suspended solids and ammonium (between 86.76% and 98.84%) were obtained in the first treatment step, mostly by indirect anodic oxidation as well as the pyrolytic degradation and oxidation with free radical species inside the cavitation bubbles. The highest removal of fluorides and phosphates was assessed during aluminum based electrocoagulation. Following the combined treatment, the concentrations of all measured parameters were significantly lower compared to the regulated values. Under the optimum treatment conditions, the removal efficiencies of color, turbidity, suspended solids, COD, BOD<sub>5</sub>, ammonium, fluorides and phosphates were 99.02%, 99.57%, 99.32%, 97.21%, 95.59%, 99.15%, 94.88% and 99.70%, respectively.

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