Electrochemical Treatment of Landfill Leachate under Cold Climate Conditions

Ambauen N.[1,*], Hallé C.[1], Meyn T.[1]

[1]Departement for Civil- and Environmental Engineering, Norwegian University of Science and Technology, S.P. Andersens Veg 5, 7491 Trondheim, Norway

*corresponding author: e-mail: noemi.ambauen@ntnu.no

Abstract
This study shows that electrochemical oxidation of landfill leachate (LL) is significantly affected by cold temperatures prevailing in Nordic climate areas. As hypothesized, the degradation of common wastewater parameters (TOC, COD) exhibited lower efficiency when low average temperature (13°C) was applied than compared to room temperature (25°C). At low temperature, 35 % of COD and 64 % of TOC were removed, compared to 69 % of COD and 74 % of TOC removal at high temperature. Electrochemical oxidation of ammonium (NH4) has also been observed, whereas only 1 % was removed at 13°C compared to 11 % at 25°C. In addition, the formation of three hazardous trihalomethanes (THMs) has been confirmed when a high current (7A) was applied, but THMs remained below the quantification limit of 3 ppb at low current (0.3 A). Even though kinetics decrease when cold temperatures are prevailing, electrochemical oxidation is considered still a suitable choice for the treatment of landfill leachate in areas with Nordic climate.

Keywords: Electrochemical oxidation, pollutant removal, organic disinfection by-products, temperature, applied current

1. Introduction

Background of this study is the introduction of more stringent regulations for landfill leachate discharge in Norway, expected to be in force by 2020. An appropriate treatment solution for organic and inorganic pollutants is therefore required. Electrochemical oxidation (EO) has been shown to be suitable treatment for LL (Panizza, Delucchi, and Sirés 2010). Anglada et al. (Anglada, Urtiaga, and Ortiz 2010) demonstrated further, that the EO for LL can be successfully extended to pilot scale. All of these studies were either carried out in the laboratory at room temperature or on site in southern Europe with high yearly average temperatures. The problem of low temperatures during EO of LL has not been addressed before, thus this study tries to fill this gap of knowledge. TOC, NH4 and COD are chosen to represent the organic, inorganic and oxidizable pollutants respectively.

2. Material and Methods

Landfill leachate is pre-treated on site (Mosjøen, Norway) with coagulation/flocculation followed by lamellae separation, in order to remove particulate matter and heavy metals. Raw LL has an average pH of 6.5 but due to pH adjustment for the flocculation process, the leachate exhibited a pH of 10-11. The collected LL was stored in the dark at 2°C.

Table 1. Characteristics of the landfill leachate from SHMIL (Mosjøen, Norway) after pre-treatment

Parameter	Pre-treated LL
COD [mgO_2/L]	51
TOC [mg/L]	21.5
sum PAH [µg/L]	0.62
BTEX [µg/L]	1.4
Bisphenol A [µg/L]	8.4
NH_4^+ [mg/L]	32.4

2.1. Experimental set-up

Bulk electrolysis experiments are carried out with an electrolytical flow cell (DiaClean, Switzerland). Both, cathode and anode are Si/BDD with an active surface area of 70 cm^2 and an inter-electrode gap of 1 mm. Galvanostatic conditions were chosen with an applied current of 0.3 and 7 A, both providing mass transfer limiting conditions. The limiting current density has been preliminarily determined to 0.127 A. The leachate (15 liter) is pumped with a flow rate of 300 L/h from the tank through the flow cell. The temperature of the LL is kept constant with a chiller (Julabo, Germany) at either low (13 °C) or high (25 °C) temperature. One experiment lasted for 4 hours.

2.2. Sample analysis

Samples were analyzed for total organic carbon (TOC) with a TOC – analyzer (Shimadzu, Germany), chemical oxygen demand (COD) and ammonium (NH4) were measured by using Hach-Lange cuvette tests. THMs were identified with a GC–MS–HS (Agilent, USA) by scanning the samples, using the NIST library for their identification.

3. Results & Discussion

3.1 Influence of temperature on removal of common wastewater parameters

The removal of TOC, COD and NH4 was significantly influenced by temperature. *Figure 1* shows the percentage removal of each parameter compared to its initial concentration. At 13 °C only 35 % of COD and 69 % of TOC were removed, compared to 64 % of COD and 74 % of TOC at 25 °C. Thus, lower temperatures imply a decrease in percentage removal of COD and TOC during bulk electrolysis. However, the TOC removal only differs by 5 % between the two temperatures tested whereas the COD removal differs by 34 %. This observation indicates that the degradation of COD to oxidized intermediates is mainly affected by a decreased temperature. TOC removal is less affected by lower temperatures since compounds contributing to TOC first must be further oxidized before complete oxidation. Looking at NH4, a similar impact of temperature was observed, 1 % and 11 % are removed at 13 °C and 25 °C respectively.

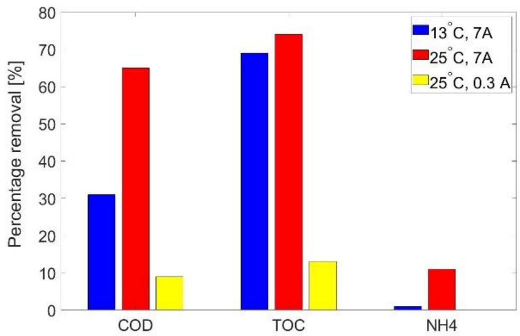

Figure 1. Percantage removal from LL at low and high temperatures.

These findings were hypothesized in advance based on the law of Arrhenius and successfully confirmed with this study. Furthermore, Kettunen et al. (Kettunen, Hoilijoki, and Rintala 1996) reported an adverse impact of low temperature (11 °C) on the anaerobic treatment of LL, yet the authors stated the process was still feaseable.

3.2 Influence of applied current on by-product formation and degradation of common wastewater parameters

Two different currents, 0.3 A and 7 A were applied at room temperature (25 °C) in order to estimate their influence on the production of THMs, a sum parameter for chlorinated organic by-products. *Figure 2* clearly depicts the formation of three THMs, trichloromethane, bromodichloromethane and dibromochloromethane when a current of 7 A is applied. However, at an applied current of 0.3 A only a marginal peak was observed for trichloromethane, which was below the quantification limit of 3 ppb. No bromated THMs were found at low current. An increase in THMs concentrations with increasing applied current was also reported by others (Anglada et al. 2011), yet they did not report a current where THMs formation could be prevented. Increasing the applied current leads to a more efficient removal of organic and inorganic pollutants than an increase in temperature (*Figure 2*). 9 % of COD and 13 % of TOC were removed at 0.3 A, compared to 65 % of COD and 74 % of TOC at 7A. No NH4 was removed at 0.3 A. However, a high current comes along with the disadvantage of promoting the formation of hazardous THMs.

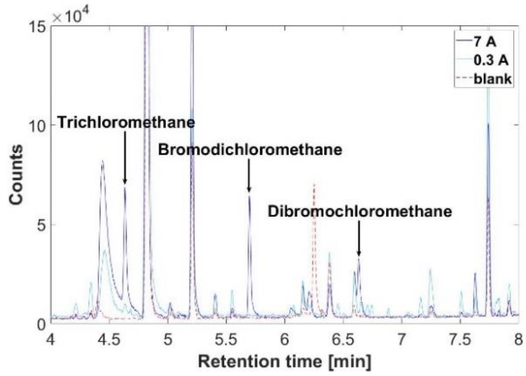

Figure 2. THMs formation at differnent applied currents

4. Conclusions

The removal efficiencies of the measured bulk parameters, COD, TOC and NH4 are all adversely affected by low temperatures. COD removal is more affected by cold temperatures than TOC removal since compounds contained in the leachate first have to undergo further oxidation before complete oxidation to CO_2 can happen. The usage of a higher current leads to a higher percentage removal of bulk parameters but comes with the tradeoff of the formation of hazardous THMs. Further investigations need to be made on the temperature effect of the formation of THMs.

References

Anglada, Ángela et al. 2011. “Boron-Doped Diamond Anodic Treatment of Landfill Leachate: Evaluation of Operating Variables and Formation of Oxidation by-Products.” *Water Research* 45(2): 828–38.

Anglada, Ángela, Ana M. Urtiaga, and Inmaculada Ortiz. 2010. “Laboratory and Pilot Plant Scale Study on the Electrochemical Oxidation of Landfill Leachate.” *Journal of Hazardous Materials* 181(1–3): 729–35.

Kettunen, R. H., T. H. Hoilijoki, and J. A. Rintala. 1996. “Anaerobic and Sequential Anaerobic-Aerobic Treatments of Municipal Landfill Leachate at Low Temperatures.” *Bioresource Technology* 58(1): 31–40.

Panizza, Marco, Marina Delucchi, and Ignasi Sirés. 2010. “Electrochemical Process for the Treatment of Landfill Leachate.” *Journal of Applied Electrochemistry* 40(10): 1721–27.