NEW PERSPECTIVE OF ELECTROCHEMICAL METHOD APPLIED TO SITE REMEDIATION: LAB SCALE STUDY FOR LEACHATE TREATMENT

Lica Carmen Georgiana[1], *Andreottola Gianni*[2], *Istrate Irina Aura*[1/2], *Rada Elena Cristina*[2], *Ragazzi Marco*[2], *Apostol Tiberiu*[1]

The aim of the research is to evaluate the effectiveness of electrochemical treatment when is applied on a leachate-contaminated soil. The electrochemical technology is usually used for wastewater treatment, but in this paper, the technology will be presented as a possible treatment for the leachate contaminated sites. The purpose of the research was to compare different tests, from economical and pollutants removal efficiency point of view. The main analyzed parameters during the development of the experiments were: the current, the initial and final pH, the values for the total and filtrated COD and the quantity of ammonia at the beginning and at the end of the tests.

Keywords: ammonia, contaminated soils, electrochemical oxidation, leachate.

1. Introduction

The transposition of the European Union legislation in the field of waste management was implemented in Romania after the entrance in the European Union. In order to comply with all the European Union regulations in the field of waste, the National Waste Management Strategy was developed by the Ministry of Environment and Water Management. This strategy was drafted for the interval 2003 – 2013, and it is to be revised on a regular basis, according to technical progress and environment protection requirements [4].In Romanian, until nowadays in landfills a high quantity of biodegradable waste can be found. At the moment at national level the highest number of contaminated sites is represented by waste disposal contaminated sites (430 not ecological landfills).

In the literature, the ammonia present in the landfill leachate represents an important problem that must be remediated because it may kill the microorganisms needed by the development of biological processes [1]; and opposite to the COD, which decreases in time (a young leachate has a concentration of COD 36 higher that an old leachate), ammonia concentrations

[1] Energy Production and Use Department, Politehnica University of Bucharest, Splaiul Independentei 313, 060032, Bucharest, Romania
[2] Civil and Environmental Engineering Department, University of Trento, via Mesiano 77, 38050 Trento, Italy

may persist in the leachate in time [3, 5], so that ammonia has been assumed as the most problematic constituent in leachate over the long term.

Taking into account all the problems that a contamination with leachate can provoke, a research activity has been performed in order to study the possibility to treat a leachate contaminated soil. The aim of this paper is to evaluate the effectiveness of electrochemical treatment when is applied on a leachate-contaminated soil. The electrochemical technology is usually used for wastewater treatment, but in this paper, the technology will be presented as a possible treatment for the leachate contaminated sites. In the experimental part, the tests were focused only on the electrochemical treatment of sand contaminated with leachate. It was chosen as solid matrix, the sand, because some previous researches have proved that the treatment is efficient on fine grain soils like clay.

2. Material and methods

Electrochemical treatment of landfill leachate was carried out in a reactor made of PVC material with a dimension of 10 cm (width) x 20 cm (length) x 10cm (height). The electrodes (anode and cathode) used for the experiments are two parallel pieces of stainless steel plates each, having a surface area of 96.4 cm^2 with a dimension of 9.8 cm x 9.8 cm. The electrodes were connected to a DC power supply in order to apply the needed voltage. The experimental setup is shown in figure 1.

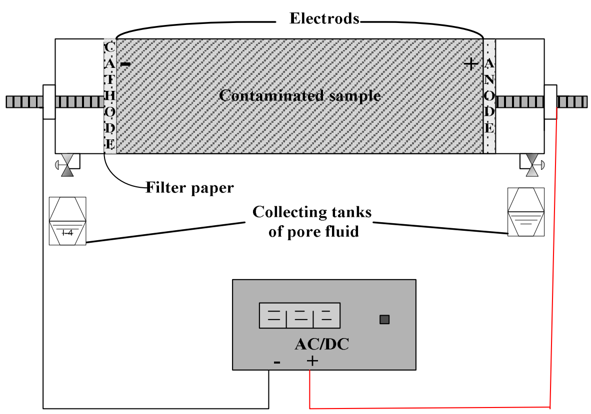

Fig. 1 The setup test used in the experimental investigation

The research has been developed on a sand sample artificially contaminated with leachate. The solid matrix that was chosen for this research is dried silica sand, named VAGA 12. From another research [2, 6], it was observed that electrochemical remediation treatment is more efficient for soils that are characterized by a very low hydraulic conductibility and a fine granulometry. The sand composition is presented in Table 1.

Table 1

Chemical composition of the sand

Compound	**Chemical formula**	**Percentages presented in soil**
Silica	SiO_2	83.3%
Iron	Fe_2O_3	2.1%
Aluminum	Al_2O_3	6.6%
Calcium	CaO	1.2%
Magnesium	MgO	1.5%
Sodium	Na_2O	2.0%
Potassium	K_2O	2.1%

The mineralogical composition of the Vaga 12 was quartz – 61.8%; granite rocks – 16.5%; feldspar – 12.7%; and 9 % other types of minerals.

The sand used for experimentation is a dissolved material, monogranulare, without particles of clay, without content of organic matter (TOC negligible amounts to 0.23g/kg_{ss}) and poor in nutrients (shortage of potassium, calcium and magnesium). This sand has also a low cation exchange capacity (CEC) of 0.7 m_{eq}/100g of soil and consequently a low power buffer.

The pH of this soil is about 8, and from the analysis it results that the sand has an iron and manganese content of about 14425.5 mg/kg_{ss}, respectively about 324.03 mg/kg_{ss}. All the main characteristics of the sand that were used during this runs are presented in Table 2.

Table 2

Chemical parameters of the sand used in the experimental research

pH	**CEC** [m_{eq}/100g]	**Fe** [mg/kg_{ss}]	**Mn** [mg/kg_{ss}]	**K_h** [m/s]	**TOC** [mg/kg_{ss}]
8.0	0.7	14425.5	324.03	10^{-4}	230

The leachate sample used to contaminate the soil, previous to the application of electrochemical treatment, is original from the Zuclo landfill situated in the north-east of Trentino county, Italy. The initial concentration of COD and N-NH_4 was 839 mg/l, respectively 140 mg/l with a pH of 7.22.

The tests performed are presented in table 3, where the main characteristics for each test (exposure time, voltage, etc.) are indicated.

Table 3

Tests performed during the present research

Test	Matrix	Contaminant	Sample weight	Sample volume	Sample length	Exposure time	Voltage (constant)	Specific voltage	pH	Initial current
			[Kg]	[L]	[cm]	[d]	[V]	[V/cm]	-	[mA]
LE_S1	Fine sand	Leachate	2.8	1.395	15.5	14	7.75	0.5	7.48	6.42
LE_S2	Fine sand	Leachate	2.8	1.395	15.5	14	15.5	1	7.48	40
LE_S3	Fine sand	Leachate	2.5	1.26	14	14	21	1.5	7.48	28

3. Results and discussion

The preparation of the contaminated sample has been developed in three phases. First of all the clean sand has been dried at 105^0C for 24 hours after which the sample has been contaminated with leachate until the sample reached saturation.

Previous to the contamination phase the original leachate was diluted with water in a ratio of 1:10 in order to have a better simulation of the real conditions in a landfill. After 14 days of treatment the sample was divided in four parts, three parts that correspond to different distances from the anode that where identified as anode, middle and cathode, and one sample identified as mixed that characterized the entire sample. It was decided such a division in order to evaluate the contaminant removal at different distances from the electrodes. The remediation results of the soil matrix are presented in table 4.

Table 4

Results of the run tests

Test		COD filtrated	N-NH4	N-NH4	COD total
		[mg/l]	[mg/l]	[%]	[%]
	Initial	14	16.5	-	
Test 1 0.5 V/cm	Anode	9	0.1	99.39	
	Middle	9	0.4	97.58	
	Cathode	5	0.2	98.79	
	Mixed	-	0.2	98.79	
Test 2 1 V/cm	Anode	-	0.7	96.70	
	Middle	-	0.3	98.58	
	Cathode	-	0.1	99.53	
	Mixed	**4**	**0.3**	**98.58**	**71.43**
Test 3 1.5 V/cm	Anode	14	0.9	94.55	
	Middle		0.8	95.15	
	Cathode		1	93.94	
	Mixed	14	0.6	96.36	

In the figures 2 and 3 the variation of ammonia removal and respectively the pH along the soil matrix are presented. It can be observe that the removal of ammonia is uniform in all the 3 tests along the matrix. The best results were obtain in the test with a specific voltage of 0.5 V/cm and 1 V/cm.

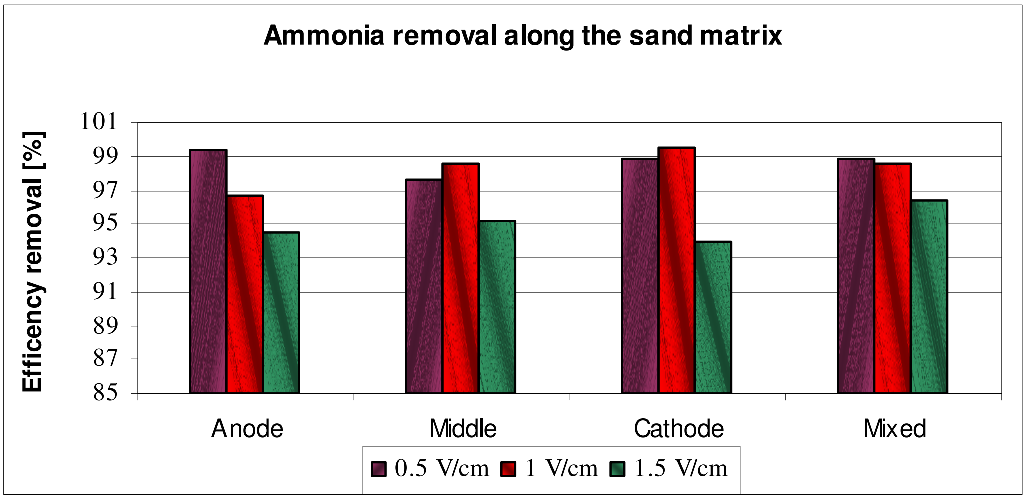

Fig. 2. The percentages of Ammonia removal along the sand matrix for the run tests

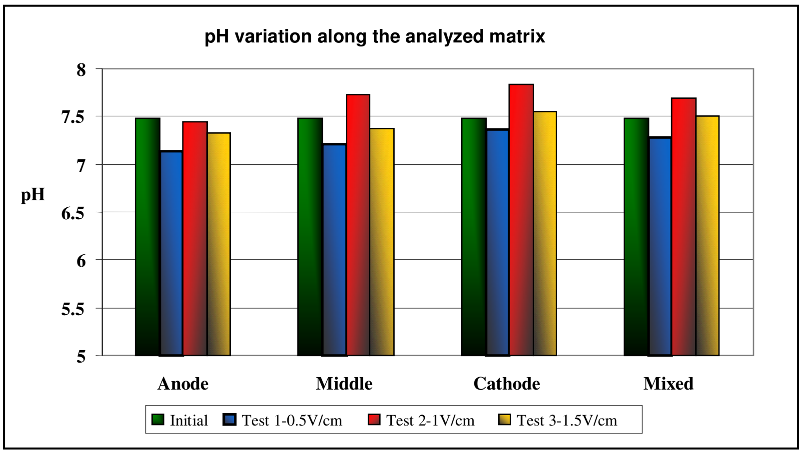

Fig. 3. The initial and the final pH for the tests run

From figure 3 it can be noticed that the soil pH drops at the anode and increases at the cathode, but the increase and the drop of pH is not so big because of the low power buffer capacity of the sand.

4. Energy consumption

From the values of voltage and current, it was possible to make an approximate estimation of the energy consumption required for the remediation action. In order to calculate the energy costs, the current consumed can be approximated with the area below the curve of the variation of the current in time. This area was calculated using AutoCad.

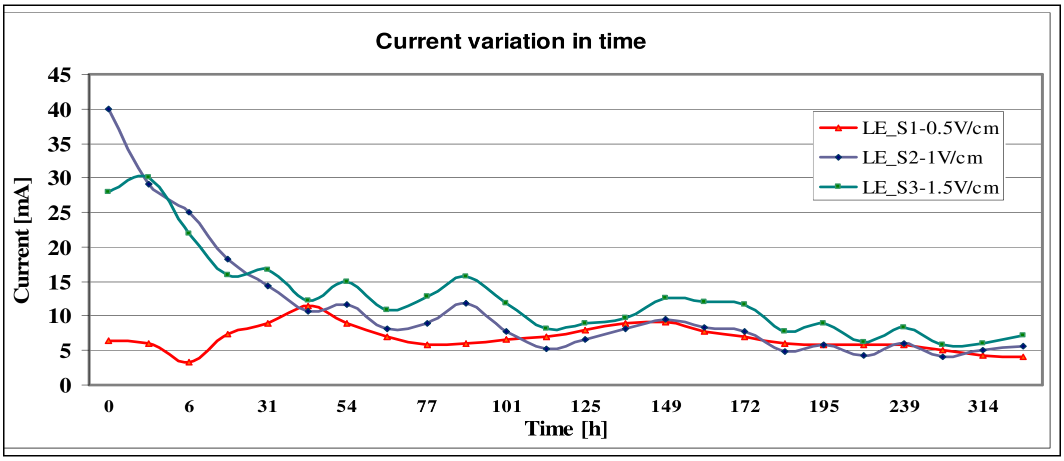

Fig. 4. The variation of the current in time for the tests with the sand matrix

For all the three tests, the energy consumed is presented below in table 5.

Table 5

Current consumption for the tests run

Test	**LE_S1**	**LE_S2**	**LE_S3**	
Matrix	Sand+Leachate			
Applied voltage	0.5	**1**	1.5	[V/cm]
Time	14	14	14	[Days]
Current consumption	**2.13**	**2.77**	**3.51**	[A]

Once the current consumption was calculated, the energy cost per unit volume (E) was calculated according the following expression:

$$E = \frac{1}{V_s} V \int * i(t) * dt \ , \qquad (1)$$

Where:

V_s is the volume of treated soil;

i is the electric current flowing;
t is time;
V is the applied voltage
$\int i(t) * dt$ is the current consumption.

The maximum power required for the test (W_{max}) was calculated as the product of the maximum current recorded (i_{max}) and of the applied voltage (V):

$$W_{\max} = i_{\max} * V, \quad (2)$$

In table 6 the energy expenditure for each test performed, is presented.

Table 6

The energy expenditure for the test performed

Test	Applied voltage	E	E	W_{max}	Total current consumption	Cost of the consumed energy[3]
	[V/cm]	[kJ/l]	[kWh/m^3]	[W]	[A]	[Euro/m^3]
LE_S1	0.5	42.50	11.80	0.071	2.13	1.3
LE_S2	**1**	**110.81**	**30.78**	**0.62**	**2.77**	**3.38**
LE_S3	1.5	210.69	58.52	0.63	3.51	6.43

For the tests performed on sand (the soil samples having a volume of about 1.25 -1.4 L and a mass about 2.5-2.8 kg) the estimated energy expenditure increased with the applied voltage, ranging from 42.5 kJ/L, corresponding to 11.8 kWh/m^3 (test LE_S1, 0.5 V/cm) to 210.7 kJ/L, corresponding to 58.53 kWh/m^3 (test LE_S3, 1.5 V/cm).The maximum applied power (corresponding to the maximum current) was about 0.07 -0.63 W . For the tests with the sand matrix the cost of the energy increases with the applied voltage.

5. Conclusions

The advantage of the electrochemical oxidation treatment applied to leachate contaminated soil is its potential for cost-effective in situ and/or ex situ use, and can be use as a pre-treatment of leachate or to remediate the contaminated soil with leachate.

The applied voltage seems to have a limited influence on the system efficiency, good results being achieved with specific voltages as low as 1 V/cm.

The tests performed at the laboratory scale showed that the removal percentages for ammonia reached almost 100% (98.6%) and for COD 71.5% for the in situ treatment of the sand matrix with an applied voltage of 1 V/cm.

[3] With a price of electricity in Romania at the end of 2008 of 0.13 [Euro/m^3]

As it is shown in the tests performed, the efficiency of ammonia removal along the sand matrix (anode, middle and cathode) is uniform (96% -99%), unlike what the literature data shows on the electrochemical oxidation treatment applied on clay, witch is recording an uneven removal along the matrix of clay with the highest removal efficiency at the anode [3].

For the tests with the sand matrix the cost of the energy increases exponential with the applied voltage. The power consumed is a key factor for assessing the economics for electrochemical treatment of landfill leachate. For the test with a specific voltage of 1V/cm, 31 kWh/m^3 was required with an energy cost for Romania of 3.38 $€/m^3$.

This treatment can be suitable for the leachate contaminated soil remediation using a specific voltage of 1V/cm, because of the high content removal of ammonia and the percentage of 71.5% removal of COD and the low energy expenditure comparing with others treatment present in the literature.

For Romanian old landfills, which the majority have to be remediated according EU, Electrochemical Oxidation could be a good treatment for the soil contaminated with leachate, because this treatment don't need the excavation of the contaminated soil and is a cost effective technology and has also good remarks on removal of ammonia and COD and also for removing heavy metals.

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