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Hybrid processes for treatment of landfill leachate: Coagulation/UF/NF-RO and adsorption/UF/NF-RO

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ABSTRACT

Landfill leachate production and management are now recognized as one of the greatest problems associated with environmentally operation of sanitary landfills. In this study, laboratory-scale experiments were implemented to assess the effectiveness of coagulation/ultrafiltration (UF) and adsorption/UF as a pretreatment option for treating stabilized landfill leachate using nanofiltration (NF) and reverse osmosis (RO). Significant number of parameters was systematically monitored: chemical oxygen demand (COD), ammonium, turbidity, pH, total organic, inorganic carbon, total organic carbon (TOC), conductivity and concentrations of anions and metals. Coagulation showed better reduction of COD (65%), TOC (86%), and turbidity (87%) than adsorption (32%, −132%, 7%, respectively). Ultrafiltration was better after adsorption since reduction of COD and TOC was higher confirming that larger molecules were removed with coagulation and smaller with adsorption. Reduction of ammonium was relatively weak, for all pretreatment steps, in amount around 15%. XLE and NF90 membranes showed better performances than NF270 membrane due to tighter structure. Removal of COD and TOC was higher than 80% while for NF270 was 37% and 73%, respectively. Initial flux of RO/NF membranes declined between 45% and 72%. However, washing returned flux to almost initial flux showing that fouling wasn't irreversible.

1. Introduction

Population on the Earth grows exponentially and changes in the consumption habits have been accompanied by the rapid generation of municipal solid wastes (MSW). Average annual waste production in Croatia, from 2005 to 2013, was 379 kg per person [1]. Average production in Zagreb, largest city in Croatia, was around 380 kg per person. One of the most common methods for managing MSW in European Union [2], as it is in Croatia, is landfilling, which generates highly polluted wastewater called landfill leachate.

Landfill leachate have been identified as an important potential contamination source of ground and surface waters, as they may percolate through soils and subsoils, causing extensive contamination of streams, creeks and water wells, if they are not properly collected, treated and safely disposed [3].

Leachate is highly variable and heterogeneous with very complex composition [4–6]. The composition and concentration of contaminants are mainly influenced by the age of the landfill [7,8]. Typically, old leachates are stabilized and characterized by relatively low chemical oxygen demand (COD), slightly basic (>7.5) and low biodegradability ($BOD_5/COD < 0.1$). These indicate that physical-chemical processes are appropriate for treatment of stabilized leachate [5,9]. In last decade nanofiltration (NF) and reverse osmosis (RO) were extensively used for treatment of landfill leachate [4,5,9] due to the stringent legal regulations. Removal efficiency of COD, ammonium and metals was up to 100% [5,9]. However, they are subjected to the flux decline, i.e. fouling [10,11]. Mainly concentration polarization contributes to membrane fouling. Therefore, membrane filtration is not suitable as a single process in landfill leachate treatment.

Hence, the aim of this study was to investigate hybrid processes for treatment of landfill leachate. Coagulation/ultrafiltration(UF)/NF-RO and adsorption/UF/NF-RO were used for treatment of landfill leachate generated in landfill located in Zagreb. One of the main goals was to use these two pretreatments in order to achieve better characteristics by NF/RO membranes (i.e. removal, low flux decrease).

2. Materials and methods

2.1. Materials

Leachate samples investigated in this study were collected from Jakuševec landfill, located in Zagreb, capital city of Croatia. It is

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located very close to the center of the city (around 14 km by air) and next to river Sava. This is an old and active landfill and produces minimum 1000 m^3 and maximum 21000 m^3 of leachate per day during summer and winter, respectively. Both raw leachates used in this study were collected from leachate basins, stored at 7 °C, and tested within 1 day of collecting the samples. Their characteristics are shown in Table 1. Leachate samples were collected two times with an interval of one week. Following parameters were systematically monitored: conductivity (κ), turbidity, COD, pH, total carbon (TC), inorganic carbon (IC), total organic carbon (TOC), ammonium (NH_4^+-N), chloride (Cl^-), fluoride (F^-), nitrite (NO_2^-), nitrate (NO_3^-), sulfate (SO_4^{2-}), bromide (Br^-), and heavy metals. From Table 1 are visible differences between two leachate samples probable because of heavy rain between two sampling campaigns.

2.2. Jar and adsorption test and procedure

Leachate sample 1 and sample 2 were used for coagulation/UF/NF-RO and adsorption/UF/NF-RO treatment, respectively.

Prior the experiments both optimal concentration of coagulant ($FeCl_3$) and optimal concentration of activated carbon (AC) were determined. Optimal coagulant dose (1 g/L of Fe^{3+}-ions) was determined as previously published by Košutić et al. [12]. Optimal concentration of AC was determined with Jar test and various weight of carbon (0.5, 1.0, 2.5 and 5.0%) during 4 h. COD and TC were monitored in 15, 30, 45, 60, 120 and 240 min. Speed of mixing was 200 rpm. Used activated carbon was in the form of stick, i.e. pellets (∅ 1.2 mm and length 3–6 mm) with specific activated surface 1145.44 m^2/g. Specific activated surface was determined with Micromeritics ASAP 2000 gas sorption analysis unit using Brunauer-Emmet-Teller method.

The adsorption was performed: (i) leachate sample was added into 1 L beaker, (ii) AC was added to the sample according to optimal concentration, (iii) solution was mixed (200 rpm for 4 h), (iv) AC settled down.

Supernatants were carefully extracted by plastic syringe about 2 cm below liquid level and were analyzed.

In order to study the effect of direct coagulation treatment of landfill leachate, no pH adjustment was made during the experiment.

After coagulation and adsorption a filtration through pure alpha cellulose filter paper (Munktell Grade 389 and 391; average pore diameter was 12–2 μm).

2.3. Membrane treatment

Used membranes were GM from GE Osmonics. Working pressure was 3 bar, flow rate 750 mL/min and surface area of the membranes 10.7 cm^2. Treatment with UF membranes lasted for 15 h until sufficient quantity of permeate was collected for RO/NF treatment. At the end membranes were cleaned.

Commercially available membranes from Dow/FilmTec (Midland, MI, USA) examined in this experiment included RO (XLE) and NF (NF90 and NF270). All membranes were stored in a dark cold place (refrigerator) before they were used. The experiment, in circulation mode, was conducted in a laboratory set-up, described in details by Dolar et al. [13], at a working pressure of 15 bar and flow rate of 750 mL/min. The surface area of the membranes was 10.7 cm^2. After the treatment membranes were washed with demineralized water and cleaned with alkaline agent (1.5% NALCO 99). The cleaning agent (temperature between 34 °C and 37 °C) was circulating for 30 min followed by soaking the membrane for 30 min in the same agent.

2.4. Hybrid processes

Schematic representation of hybrid processes for treatment of landfill leachates is presented in Fig. 1. Samples were systematically monitored during whole hybrid process in sampling points presented by red dots.

2.5. Analytical methods

Conductivity was determined by conductometer (SCHOTT Instruments Lab 960, Germany). TOC, IC and TC were determined

Table 1
Characterization of Jakuševec landfill leachate.

Parameter	Concentration[a] (mg/L)		Parameter	Concentration[a] (mg/L)	
	Sample 1	Sample 2		Sample 1	Sample 2
Color	Very dark brown		Co	4.19×10^{-2}	2.60×10^{-2}
COD (S.D.)	1380 (34)	747 (58)	Cr	1.64×10^{-1}	9.62×10^{-2}
NH_4^+-N	665.2	307.4	Cu	2.45×10^{-2}	1.33×10^{-2}
Turbidity/NTU	100	48.4	Fe	3.88	2.36
pH	7.94	7.68	K	525	338
TC	1993.0	891.7	Mg	84.2	5.77
IC	828.4	476.2	Mn	4.01×10^{-1}	3.77×10^{-1}
TOC	1164.6	415.5	Na	7.62×10^{-4}	5.27×10^{-4}
κ/μS/cm	8630	5830	P	2.14	$<1.0 \times 10^{-7}$
F^-	5.03	4.54	Pb	4.40×10^{-2}	8.33×10^{-3}
Cl^-	10483.84	6754.93	Pt	1.01×10^{-4}	6.11×10^{-5}
NO_2^-	1390.60	1012.64	Rb	3.12×10^{-1}	2.10×10^{-1}
NO_3^-	119.91	79.24	Se	1.65×10^{-2}	9.06×10^{-3}
SO_4^{2-}	199.26	479.64	Si	12.6	8.09
Br^-	7.60	15.10	Sn	2.26×10^{-2}	1.31×10^{-2}
Ag	4.39×10^{-3}	1.50×10^{-4}	Sr	6.87×10^{-1}	5.49×10^{-1}
Al	3.67×10^{-1}	1.43×10^{-1}	Th	$<1.0 \times 10^{-7}$	$<1.0 \times 10^{-7}$
As	3.44×10^{-2}	2.72×10^{-2}	Ti	2.38×10^{-1}	2.15×10^{-1}
Au	3.18×10^{-3}	2.19×10^{-3}	U	$<1.0 \times 10^{-7}$	$<1.0 \times 10^{-7}$
B	6.74	5.20	V	4.52×10^{-2}	2.93×10^{-2}
Bi	118	2.39×10^{-1}	W	6.23×10^{-3}	4.13×10^{-3}
Ca	128	107	Zn	1.90	7.05×10^{-2}
Cd	4.40×10^{-4}	6.55×10^{-5}			

S.D. – standard deviation (N = 3).
[a] Concentrations are in mg/L or otherwise written.