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Influence of Leachate Quality on Reverse Osmosis Performance

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1. Introduction

Landfills are still the most popular method for municipal waste disposal. One of the main environmental problems arising from solid waste landfilling is the generation of landfill leachate from original moisture in solid waste and percolation of rainwater through the final cover soil (Theepharaksapan et al. 2011). Production of landfill leachate begins with introducing moistured waste into disposal area and continues for several decades following the landfill closure (Hasar et al. 2009). The chemical and microbiological composition of landfill leachate is complex and variable, since apart from being dependent upon features of residual deposit, it is influenced by environmental conditions, the operational manner of the landfill and by dynamics of the decomposition process that occurs inside the cells (Yao 2017, El-Fadel et al. 2002, Kjeldsen et al. 2002). According to Christensen et al. (2001) and Moravia et al. (2013) one of characteristic features of leachate is presence of four groups of pollutants: dissolved organic matter, macro inorganic compounds, heavy metals and xenobiotic organic compounds originating from chemical and domestic residue present at low concentrations (aromatic hydrocarbons, phenols, pesticides, etc.) and microorganisms. Since the composition of leachate consists of a wide range contaminants, it can be toxic for environmental and human health (Hasar et al. 2009). For this reason, leachate treatment is a major issue in the context of landfill management and treating design should take into account not only quantity and quality but also variation over time, in line mainly with landfill age (Morello et al.

2016). For young leachate (from landfills under 5 years of exploitation), which contains mostly organic substances in biodegradable form, nutrients and suspended solids, a conventional biological process could be effective (Rukapan et al. 2012, Theepharaksapan et al. 2011, Yahmed et al. 2009, Nowak et al. 2016). For old landfills, most of the leached organic compounds are hardly- or non-biodegradable forms and such a leachate should be treated by physico-chemical processes or by a combination of biological and physico-chemical processes (Fudala-Książek et al. 2016, Rukapan et al. 2012). Among another process, the reverse osmosis (RO) seems to be one of the most promising and efficient method for landfill leachate treatment. The application of RO for treatment of stabilized leachate from Lipówka landfill (Poland) gave for COD a 97% removal efficiency (Peters 1998). During the treatment of stabilized leachate from Hedekosga (Sweden), the maximum removal of COD and $N\text{-}NH_4^+$ with initial concentrations of 1,254 and 541 mg/l was found to be 95 and 82%, respectively (Thörneby et al. 2003). Reverse osmosis was also employed for the treatment of leachate from Wijsler landfill (The Netherlands) The reduction of COD and $N\text{-}NH_4^+$ was found to be 98% with the initial concentrations of 335 and 140 mg/l, respectively (Kurniawan et al. 2006).

Even though literature review indicates that leachate treatment by RO process is well studied topic, there are no wider and systematic investigations and comparison on the RO performance for young and stabilized leachate. Fouling of reverse osmosis membrane, during leachate treatment, differs for young and stabilized leachate due to the fact, that chemical composition of these leachate differs from each other. The differences in RO performance influence on efficiency of reverse osmosis system. Besides, most of the papers describe the efficiency of RO systems based on laboratory tests conducted on lower flows and lower pressures, which does not reflect the actual operating conditions of the system (Hasar et al. 2009, Theepharaksapan et al. 2011, Rukapan et al. 2015). In addition, most studies are conducted on easy-to-use flat membranes while in the actual leachate treatment conditions most commonly recommended are tubular membranes with parallel flow (Bohdziewicz et al. 2010, Wang et al. 2014). Therefore the main objective of this work is to evaluate the influence the two kind of leachate (young and stabilized) on reverse osmosis performance during their treatment by the use of the

RO system which reflects the actual operation circumstances of reverse osmosis units exploited on municipal landfills.

The use of reverse osmosis for landfill leachate treatment may involve various difficulties associated with both the nature of the leachate (variable quantity and quality over time) and the specificity of the reverse osmosis process (scaling, fouling, adsorption on the membrane surface). Therefore realization of this study is of practical importance to the operator of the leachate treatment plant, because it will evaluate the effectiveness of the unit reverse osmosis process in the treatment of landfill leachates and validity of its use for both young and stabilized leachate.

2. Materials and methods

The leachate samples used in this study were collected directly from the municipal landfill site localized in northern-eastern part of Poland, at the northern latitude 53°50’55’’ and the eastern longitude 22°19’02’’. The young leachate (ON) was taken from landfill cell, which is exploited since three years. The mature stabilized leachate (OS) was taken from landfill cell with over 20 years of exploitation. The samples both of young and mature leachate were collected three times. Collected samples were transported to the laboratory and stored in dark at 4°C to minimize biological and chemical changes.

In leachate samples the following parameters were determined: electro-conductivity (EC), chemical oxygen demand (COD), biological oxygen demand (BOD), nitrogen ammonia N-NH_4; total nitrogen (TN), total phosphorus (TP), sulfate (SO_4^{2-}), chloride (Cl^-), calcium (Ca^{2+}) and manganese (Mg^{2+}). All determination were done in accordance with APHA (APHA 1995).

The evaluation of the reverse osmosis system operating parameters was carried out separately for young leachate and stabilized leachate. The reverse osmosis process was carried out using the RO20NS.1 – a laboratory reverse osmosis system, which was designed and constructed in such a way as to reflect the actual operation of reverse osmosis units running in municipal waste landfills. The basic elements of the laboratory system were the Micro 240 PCI membrane module and the high pressure pump. In addition, the system was equipped with a pump pulsation damper, a flow meter, safety valve and two pressure gauges enabling pressure measurement on the liquid entry side to the system and to control the RO process pressure. The leachate treatment process was carried out in a cross-flow system with concentrate recirculation to the tank supplying the effluents to the RO system. The recovery rate was set at 60/40 (60% permeate and 40% concentrate). Before entering the RO system, landfill leachate were acidified to pH 6.5 and subjected to filtration successively on a 50 and 5 µm filter. The RO process was carried out at the feed rate of 18 dm^3/min, 25 ° C and constant pressure of 3.8 MPa. The scheme of research installation is given in Figure 1.

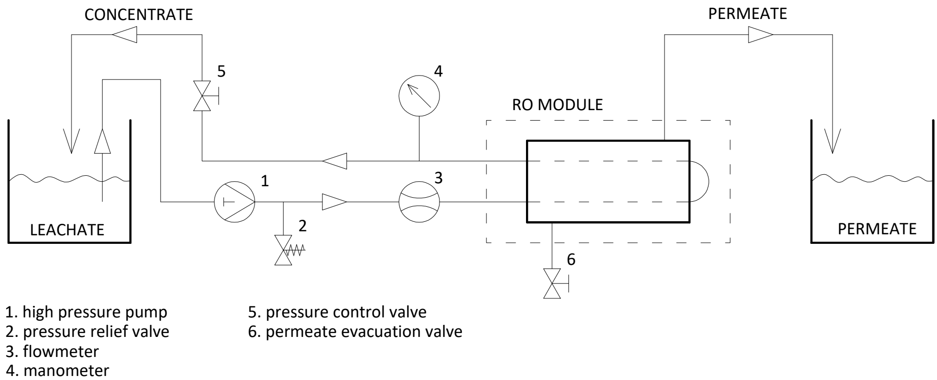


Fig. 1. Scheme of RO installation
Rys.1. Schemat instalacji RO

The performance characteristics of the reverse osmosis system were made on the basis of the following calculations:

- permeate flux (J):

$$J = k \cdot \left(\frac{V_p}{t \cdot S}\right) \quad (\mathrm{L/m^2 \cdot h}) \tag{1}$$

where: V_p – permeate volume (L), t – time (h), S – membrane area (m^2), k – temperature correction factor;

- removal (retention) rate of salt expressed as a electroconductivity value (R_{EC}):

$$R_{EC} = \left(1 - \frac{EC_p}{EC_n}\right) \cdot 100\% \quad (\%) \tag{2}$$

where: EC_p – electroconductivity of permeate (mS/cm),
EC_n – electroconductivity of inlet (leachate) (mS/cm);

- concentration factor of EC in concentrate (CF_{EC}):

$$CF_{EC} = \frac{EC_c}{EC_n} \tag{3}$$

where: EC_c – electroconductivity of concentrate (mS/cm).

In order to assess the relationship between the quality of leachate directed to the RO system and its operating parameters, the analysis of variance (ANOVA) was carried out. The one-way ANOVA with one independent variable: kind of leachate (with two levels: young leachate, stabilized leachate) and 5 dependent variables (filtration time, J_{min}, J_{aver}, J_{max}, R_{EC}) was used for test. To facilitate the interpretation of the results, the average, minimum and maximum permeate flux during reverse osmosis process was also calculated.

3. Results and discussion

Table 1 presents the basic characteristics of young and stabilized leachate directed to the reverse osmosis system.

The leachate from the new disposal field (ON) are characterized by more than three times higher COD value and six times higher value of BOD compared to stabilized leachate (OS). This indicates a high share in the leachate of readily biodegradable organic substance with high molar mass (Rosik-Dulewska 2002). Younger leachate also had higher concentrations of total and ammonia nitrogen as well as total phosphorus as well as a higher value of conductivity. In both effluents, the concentration of magnesium and calcium ions, chlorides, sulphate and iron is maintained at a similar level.

The collected leachate samples were directed to the reverse osmosis system, with the osmotic filtration being carried out in three series, separately for young and stabilized leachate. The results obtained are shown in Figure 2 and Table 2.

Average permeate flux for stabilized leachate was 38.61 $L/m^2 \cdot h$, and the time required to achieve the assumed 60% recovery rate was

5.27 h (Table 2). The biggest decrease in permeate flux occurred in the first hour of the system's operation, indicating the membrane blocking. In the final phase of the process permeate flux was on average 28.83 $L/m^2 \cdot h$ (50% of maximum permeate flux). At the 60% recovery rate, the EC concentration factor was reached at 2.01, which means that at 2.5 times the reduced volume of the concentrate, a two-fold increase in the electrolytic conductivity was achieved. The average rate of EC removal was 99.1%.

Table 1. Basic parameters of young (ON) and stabilized (OS) leachate directed to the reverse osmosis system
Tabela 1. Podstawowe parametry odcieku młodego (ON) i ustabilizowanego (OS) kierowanego na układ odwróconej osmozy

Parameter	Young leachate (ON) (mg/L)		Stabilized leachate (OS) (mg/L)	
	Average	Stand. dev.	Average	Stand. dev.
EC	15.37	2.73	7.89	3.71
COD	4623.3	1985.6	1443.3	1782.3
BOD	579.3	191.7	92.0	70.2
$N\text{-}NH_4^+$	763.3	127.4	231.7	145.8
TN	826.7	176.2	275.0	152.1
TP	16.13	1.62	0.82	1.04
SO_4^{2-}	235.7	152.5	376.9	278.0
Fe	2.73	1.97	4.30	1.75
Cl^-	1959.3	469.9	1388.8	825.6
Mg^{2+}	387.5	120.9	376.7	135.3
Ca^{2+}	469.5	50.2	446.8	23.7

All in mg/dm^3 apart EC (mS/cm)

In the case of young leachate, the filtration time required to achieve the 60% recovery rate was longer and amounted to nearly 7 h, which was the result of a lower average permeate flux – 29.35 $L/m^2 \cdot h$. As in the·case of stabilized leachate, the largest drop in flux occurred in the first hour of filtration. However, the observed drop in permeate flux during filtration of young leachate was much larger; the final value of permeate flux was on average 19.39 $L/m^2 \cdot h$ (29% of maximum permeate flux). The value of concentration factor was 1.97 and the EC removal rate – 98.8%.

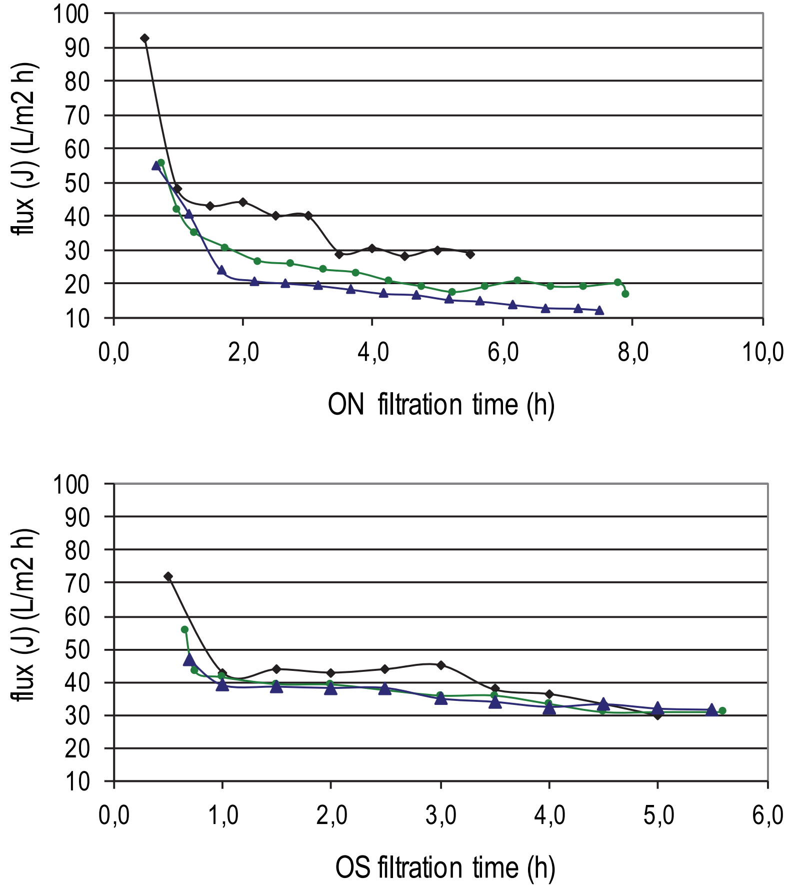


Fig. 2. The value of permeate flux during young (ON) and stabilized (OS) leachate filtration in reverse osmosis process

Rys. 2. Przepływ permeatu podczas filtracji odcieku młodego (ON) I ustabilizowanego (OS) w procesie odwróconej osmozy

Table 2. Characteristic of membrane performance according to kind of leachate
Tabela 2. Parametry pracy membrany w zależności od rodzaju kierowanych na nią odcieków

Parameter		Stabilized leachate (OS)	Young leachate (ON)	Average for all groups
		N = 3	N = 3	N = 6
Filtration time t (h)	Average	5.27	6.97	6.12
	Stand. dev.	0.49	1.29	1.28
Flux minimum J_{min} ($L/m^2 \cdot h$)	Average	28.83	19.39	24.11
	Stand. dev.	4.21	8.58	7.95
Flux average J_{aver} ($L/m^2 \cdot h$)	Average	38.61	29.35	33.98
	Stand. dev.	3.25	10.60	8.65
Flux maximum J_{max} ($L/m^2 \cdot h$)	Average	58.05	67.82	62.94
	Stand. dev.	12.95	21.63	16.82
EC removal rate R_{EC} (%)	Average	99.1	98.6	98.8
	Stand. dev.	0.26	0.40	0.40
concentration factor CF	Average	2.01	1.97	1.99
	Stand. dev.	0.11	0.02	0.07

The main reason for the drop in permeate flux, observed both during osmotic filtration of young and stabilized leachate, was the process of fouling/scaling and the increase in osmotic pressure, caused by the increasing concentration difference between permeate and leachate/concentrate mix (Costa and Pinho 2005). Kabsch-Korbutowicz and Majewska-Nowak (1996) explain increased membrane blocking during filtration of young leachates by humic substances present in leachate. Their polyelectric nature facilitates binding of cations by these compounds (including Ca^{2+}, Mg^{2+}), which neutralizes their negative charge resulting from the deprotonation of H^{+} ions and facilitates blocking of pores in negatively charged membrane. The carried out research shows that an important role in the neutralization of the charge of humic substances can also be played by $N\text{-}NH_4^{+}$ ions. Formed in this way complexes of inorganic ion with humic substances have, in addition to a neutral electric charge, a smaller solubility than free humic macromolecules, hence the polarization layer is formed faster at the membrane surface (Kabsch-Korbutowicz and Majewska-Nowak, 1996). For this reason, the filtration of solutions containing humic substances and inorganic cations causes a decrease in membrane permeability. In addition, surface membrane studies carried out by Chon et al. (2012) indicated that N-NH_4^+ along with Fe^{3+}, Al^{3+}, Si^{4+} and Ca^{2+} are the main inorganic foulants on RO membranes. The conducted research indicates that the process of filtration of stabilized leachate is characterized by higher flow rates and smaller values of flux decline. When RO system receives leachate rich in humic substances, the removal of ammonium cations from it may bring beneficial effects in the form of reducing the membrane blocking and increasing value of permeate flux.

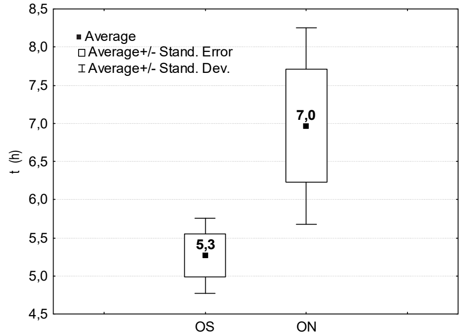
In order to assess the effect of leachate age on RO system performance, the analysis of variance (Tab. 3) and average values of RO system parameters were conducted (Tab. 2, Fig. 3).

Although the analysis of variance indicated the lack of statistically significant differences between the parameters of the reverse osmosis system work during filtration of young and stabilized leachates (Tab. 3), the analysis of average parameters of the system performance indicated a shorter filtration time – by more than one hour - of stabilized leachate, higher average permeate flux, higher EC removal rate and better concentration factor. The observed differences indicate that the filtration of stabilized leachate affects the better performance of the RO system compared to filtration of young leachate (Tab. 3, Fig. 3).

Table 3. Influence of kind (age) of leachate on reverse osmosis performance – variance analysis
Tabela 3. Wpływ rodzaju (wieku) odcieków na parametry pracy odwróconej osmozy – analiza wariancji

Parameter	SS Effect	df Effect	MS Effect	SS Error	df Error	MS Error	F	p
Filtration time t (min)	4.33	1	4.33	3.8	4.0	0.9	4.6	0.10
J_{min} ($L/m^2 \cdot h$)	133.7	1	133.7	182.6	4.0	45.7	2.9	0.16
J_{aver} ($L/m^2 \cdot h$)	128.8	1	128.8	245.6	4.0	61.4	2.1	0.22
J_{max} ($L/m^2 \cdot h$)	143.1	1	143.1	1271	4.0	317.8	0.5	0.54
EC removal rate (%)	0.350	1	0.350	0.5	4.0	0.1	3.1	0.16

SS Effect – sum of squares between groups, MS Effect – mean square between groups, SS Error – sum of squares inside group, MS Error – mean sum of squares inside group, df – degrees of freedom, F – value of Test F, p – probability level



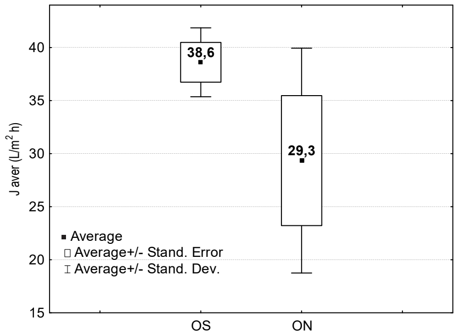


Fig. 3. Filtration time (t) and permeate average flux (J_{aver}) according to the kind of leachate
Rys. 3. Czas filtracji (h) i średni przepływ permeatu (J_{aver}) w zależności od rodzaju odcieków

4. Conclusions

Osmotic filtration of young leachate with high organic content is characterized by lower permeate flux rates and higher flux decline in comparison with stabilized leachate. Directing leachate from old disposal fields to the RO system caused an increase in the permeate flux (J aver, J min) and shortening the leachate filtration time. Higher ionic strength, characteristic for leachate from young landfills (ON), tends to block the pores of the membrane. In addition, the rate of salt retention is lower then. Moreover, high organic content in the leachate from young landfills, with a high concentration of cations, leads to an unfavorable phenomenon of neutralization of the membrane surface. The lower concentration of cations in the feed (including $N\text{-}NH_4^+$) limits the process of neutralization of humic substances. The humic substances not neutralized by cations, maintain their negative electric charge and do not deposit on the negatively charged surface of the membrane.

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Wpływ jakości odcieków na parametry pracy odwróconej osmozy

Streszczenie

W pracy przedstawiono wpływ odcieków ze składowisk o różnym czasie eksploatacji na parametry pracy odwróconej osmozy podczas ich oczyszczania. Do badań przyjęto nieustabilizowane odcieki ze składowiska o 2-letnim czasie eksploatacji oraz odcieki ustabilizowane ze składowiska o czasie eksploatacji przekraczającym 10 lat. Analizy prowadzono na układzie odwróconej osmozy, skonstruowanym w ten sposób by odzwierciedlać rzeczywiste warunki pracy układów eksploatowanych na składowiskach odpadów. Odcieki skierowano na układ RO zachowując podczas ich filtracji te same parametry pracy (temperatura, ciśnienie, przepływ, stopień odzysku oraz odczyn odcieków). Uzyskane wyniki wskazały, że proces filtracji odcieków ustabilizowanych charakteryzuje się lepszymi parametrami pracy układu RO, tj. krótszy czas filtracji, wyższy przepływ permeatu oraz wyższy stopień retencji soli. Większa zawartość substancji organicznych zawartych w odciekach młodych (nieustabilizowanych) przyczynia się do blokowania porów membrany i obniża wydajność układu osmotycznego. Ponadto wyższa zawartość substancji organicznej w odciekach z młodych składowiska oraz wyższe stężenie w nich kationów prowadzi do niekorzystnego zjawiska neutralizacji powierzchni membrany.

Abstract

In paper an influence of leachate from landfills with different exploitation time on reverse osmosis performance during their treatment was presented. An unstabilized leachate from landfill of two years exploitation and stabilized leachate from landfill with operation time over 10 years were taken to analysis. The analyses were conducted with use of RO system constructed to reflect the actual operating conditions of the reverse osmosis system exploited on municipal landfills. These two kind of leachate was directed to the reverse osmosis system by maintaining during their filtration the same operating parameters (temperature, pressure, flow rate, recovery rate and pH of leachate). The obtained results indicated that filtration of stabilized leachate is characterized by better performance of the RO system, i.e. shorter filtration time, higher permeate flow and higher salt retention. The higher concentration of organic matter in unstabilized leachate contributes to membrane pores blocking decreases the efficiency of the osmotic system. Moreover, high organic content in the leachate from young landfills, with a high concentration of cations, leads to an unfavorable phenomenon of neutralization of the membrane surface.

Słowa kluczowe:
oczyszczanie odcieków, składowisko odpadów, odwrócona osmoza

Keywords:
leachate treatment, municipal landfill, reverse osmosis