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Performance and efficiency of reverse osmosis process in treatment of young and matured landfill leachate

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ABSTRACT

The consequence of waste landfilling is the formation of leachate characterized by varying quality and quantity. Although studies on the treatment of leachates have been ongoing for many years, the development of an efficient and effective treatment method remains an open topic. The main goal of this study was to analyze the performance and efficiency of reverse osmosis (RO) in treating young and stabilized landfill leachate. To achieve the purpose of the work, the samples of each kind of leachate were collected from municipal landfills in north-eastern Poland. Then both young and matured leachate were directed into the RO system for treatment. In raw and treated leachate (permeate) samples following parameters were analyzed: pH, electroconductivity (EC), total dissolved solids, chemical oxygen demand, nitrogen ammonia, total phosphorus, sulfate, iron, chloride, total organic carbon, manganese, and calcium. The performance of the RO system was analyzed based on permeate flux, concentration factor of EC in concentrate and recovery rate. The conducted analysis showed that the RO process displayed better performance for stabilized leachate. Permeate flux during matured leachate filtration was 38.8 L/m^2 h, while during filtration of young leachate – 29.3 L/m^2 h. The assumed recovery rate was achieved after 4.5 and 7.0 h, respectively for stabilized and young leachate. The efficiency of purification of young and stabilized leachate by RO was similar: the highest removal rate of over 99% was achieved for Fe and Cl^- and over 98% for EC, both for young and matured leachate. The lowest efficiency of purification was noted for sulfate: 88.6% and 83.2%, respectively for young and matured leachate. Considering the operation time of the RO system, the final efficiency of the treatment was achieved faster in the case of the filtration of stabilized leachates.

Keywords: Efficiency; Landfill leachate; Performance; Reverse osmosis

1. Introduction

Leachates are an inseparable element of landfill exploitation. They are generated during landfill exploitation, as well as after its closure [1,2]. The composition of leachates and the concentration of pollutants in them depends on many factors, such as type of deposited waste, the method of landfill operation, landfill age, climate, and hydrological conditions [3,4].

Over time, the generated leachate goes through several phases, in which their composition is modified due to changes in bacterial activity and conditions in the waste body [5,6]. The most pronounced differences are observed between the acidic phase, which is characteristic for landfills with less than 5 years of exploitation, and a stable methane phase observed in landfills operating for more than 10 years. In the acidic phase, the leachate is characterized by a high content of easily decomposable organic matter,

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which promotes a high value of biological oxygen demand (BOD). Consequently, it gives a value of BOD/chemical oxygen demand (COD) ratio in the range of 0.5–1.0 [7]. With time, the BOD value decreases and the COD value stabilizes. In landfills, with a long time of exploitation, the value of the BOD/COD ratio is below 0.2. The degradation of humic substances, containing organic nitrogen leads to an increase in the concentration of ammonium nitrogen, which is released as a result of deamination and ammonification processes [7,8]. The matured leachate is also characterized by a low content of total phosphorus (TP) [9]. In the acid phase at the lower pH, value the highest concentrations of heavy metals are observed in leachate [10]. In stabilized landfills, the concentration of metals in leachate is lower as a result of their reduction and precipitation as sulfide, carbonate, and hydroxides [11]. This is also confirmed by research conducted at Polish landfills in Sianów, Wysieka and Hryniewicze [12–14].

Variability in leachate quantity and quality makes the selection of an appropriate treatment system a very complex issue. For the young leachate treatment, with a high content of organic matter, the biological methods are recommended [15]. However, to remove the refractory components from the matured leachate, physicochemical processes are most frequently used [8,16–19]. One of the methods which are frequently applied in the last decade for the treatment of both young and stabilized leachates is reverse osmosis (RO) [20]. The process can be used to remove both organic and inorganic compounds [21]. Due to the versatility of the RO, it becomes a treatment system, the use of which is considered by many landfills in the country. The first RO installations in Poland were built on the landfills in Łężyce near Gdańsk (2005), Sobuczyna near Częstochowa (2008), Stary Las near Gdańsk (2012), Siedliska near Ełk (2012), Kozodrza near Ostrów, Lipówka near Starogard Gdański. Despite the many advantages offered by the RO technique, such as the very high efficiency of removing contaminants, minimal use of chemicals or the possibility of treating many types of wastewater, its application is also associated with some difficulties. The most frequent problems connected with the use of the RO for landfill leachate treatment are: formation of a biological layer on the membrane, particle deposition on the membrane surface (i.e. fouling and biofouling) and scaling. Fouling and scaling are caused by the presence of organic and inorganic matter, which may affect the removal of low molecular mass organic micropollutants [22]. These phenomena have an impact on the rate of permeate flux and its change over time [23].

The literature review shows that numerous studies on the treatment of landfill leachate using RO have been carried out worldwide in recent years [2,24–31]. The treatment of young leachate from Chung Nam landfill in South Korea was carried out using an RO system and about 96%–97% removal of COD and $N–NH_4^+$ was achieved [32]. With the system capacity of 500 m^3/d, an RO system used in Changshengqiao landfill was able to achieve 99.9% removal efficiency for ions such as Ca^{2+}, Mg^{2+} [33]. Rejection coefficients of COD and heavy metal higher than 0.98 and 0.99, respectively during pilot-scale high-pressure filtration were reported by Chianese et al. [34]. Bohdziewicz and Kwarciak [30] achieved a 98.9% removal for COD and 91.4% for BOD using the system UASB-RO in the treatment of landfill leachate from Sobuczyna in Poland. Although the issue of leachate treatment by use of RO is present in the literature, there is a niche concerning comparative analyses of performance and efficiency of the RO system in treating leachate both from young and matured landfills. In the context of this information, the main goal of this study was to analyze the performance and efficiency of RO in treating young and matured landfill leachate. The implementation of the assumed goal will contribute to increasing the knowledge on the effectiveness of using RO with reference to young and matured leachate and give more detailed data concerning RO process efficiency in dependence on the leachate maturity/age.

2. Material and methods

2.1. Material

The samples of leachate used in this study were obtained from the landfill located in Warminsko-Mazurskie Province in Poland (N53°50'49'', E22°19'05''). The landfill is operated since 1983 and has an area of 25.5 ha, including one already closed quarter and a single active one. The annual amount of waste deposited at the active part of the landfill is 20,000 mg, that is, about 50 ÷ 70 mg/d. The landfill site is properly organized from a technical and operational point of view and has an infrastructure including leachate collection facilities. The average daily amount of leachate produced on both cells varies from 50 to 90 m^3. Two types of leachate were collected for analysis: the young leachate (NEW) was taken from the landfill quarter, which is exploited for four years. The mature stabilized leachate (OLD) was taken from a closed landfill quarter with over 20 years of exploitation. The leachate samples were collected directly from the well on the drainage system. The samples young and matured leachate were collected three times. Collected samples were transported to the laboratory, stored in dark at 4°C and analyzed within 1 d from sampling. Then both of young and matured leachate were directed to the RO system for treatment.

In raw and treated leachate (permeate) samples following parameters were analyzed: pH, electroconductivity (EC), total dissolved solids (TDS), COD, nitrogen ammonia ($N–NH_4^+$), TP, sulfate (SO_4^{-2}), iron (Fe), chloride (Cl^-), total organic carbon (TOC), manganese (Mg^{2+}) and calcium (Ca^{2+}).

The EC and the pH were measured on-site by conductivity and potentiometric method, respectively, using a portable pH meter (HACH HQ40, Hach Company USA). TDS were determined by a mass balance method after a well-mixed sample filtration through a FILTRAK cellulose fiber filter. The COD was analyzed using a calorimetric method with a HACH spectrophotometer after a 2 h reactor digestion (a $K_2Cr_2O_7$ method). Nitrogen ammonia was analyzed with the use of a UV spectrophotometer. For determination of chloride, sulfates and ferrous a HACH spectrophotometer was used. The concentration of TOC was determined by ICE 3400 *atomic absorption spectrometer*, while manganese and calcium by ion chromatograph Thermo Scientific ICS 5000 (Thermo Fisher Scientific Company, USA). All parameters were analyzed according to standard methods for the examination of water and wastewater [35]. The obtained results were the mean value of three determinations carried out simultaneously.

2.2. Methods

The RO tests were carried out using a laboratory RO system, which was designed and constructed in such a way as to reflect the actual operation of RO units exploited at municipal landfills. The observation of RO performance was done separately for young leachate and stabilized leachate. The RO plan was equipped with two tubular membranes (240 PCI membrane) with a total area of 240 cm^2. Membranes were made from polyamide and characterize medium hydrophilicity and NaCl retention of 99%. Membranes were adapted to work in a high pH range (1.5 ÷ 12.0) and maximum operating pressure of max. 6.4 MPa. 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%. The system was operated with a feed rate of 18 L/min, a temperature of 25°C and a constant pressure of 3.8 MPa. The scheme of research installation is given in Fig. 1. Before entering the RO system, landfill leachate was acidified to pH 6.5 and subjected to filtration successively on a 50 and 5 μm filters.

The performance of the RO system was analyzed based on the permeate flux and recovery rate. The permeate flux (*J*) was calculated according to equation [36]:

$$J = k\left(\frac{V_p}{tS}\right)\left(\text{L}/\text{m}^2\ \text{h}\right) \quad (1)$$

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

The value of the recovery rate (Y) was obtained from the following equation [36]:

$$Y = \frac{V_p}{V_n} \times 100\% \quad (2)$$

where V_p – the volume of permeate (L), V_n – the initial volume of the inlet (leachate) (L).

The RO efficiency was calculated using the rejection coefficient (R), named also as a removal rate, which is calculated using the equation [36]:

$$R = 1 - \frac{\text{Cp}_i}{\text{Cn}_i} \quad (3)$$

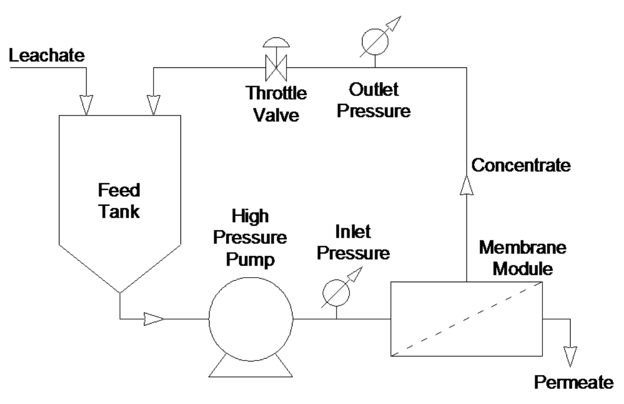

Fig. 1. Scheme of RO installation.

where Cp_i – concentration of solute i in permeate, Cn_i – concentration of solute i in the inlet (leachate).

For data analysis, Statistica software was used in this study. The basic statistics analysis included calculation of mean value and standard deviation. A parametric independent t-Student test was employed to check the statistical differences between the average RO removal rate for new and old leachate. For the statistical analysis, significance was evaluated at a probability level $p < 0.05$.

3. Results and discussion

3.1. Leachate characterization

The chemical and physical characteristics of landfill leachate are listed in Table 1. The pH value for young (NEW) leachate was 6.5 and for matured (OLD) leachate – 7.7 what is in agreement with the postulate that the pH of leachate increases with landfill age [12]. Young leachate contained much more organic substances expressed as a COD and TOC: 4,211 mg O_2/L and 9,227 mg/L in comparison to matured leachate with 1,403 mg O_2/L of COD and 954 mg/L of TOC. The BOD/COD ratio for OLD leachate was 0.1, while for NEW leachate – 0.5, suggesting that stabilized leachate was much less biodegradable than the NEW one. The concentration of nitrogen ammonia, which according to Theepharaksapan et al. [16] is identified as one of the major toxic compounds present in landfill leachate was similar for both analyzed leachates: 753 mg/L and 732 mg/L for NEW and OLD leachate. The NEW leachate contained almost two times higher concentrations of TDS and chloride. The content of manganese and calcium was similar in NEW and OLD leachate and oscillated within the limit of 400 mg/L.

The leachate characteristics did not meet the standard limits specified by the Polish Minister of Environment concerning the conditions to be fulfilled for the introduction of wastewater into water or land, and on substances that are particularly harmful to the aquatic environment (Journal of Laws of 2014.12.16). The limit values were exceeded for N–NH_4^+, COD, Cl, TOC in case of NEW and OLD leachate and for TP in case of NEW leachate.

3.2. RO performance and efficiency

Leachate from new and old, closed landfill quarter, after their acidification (if necessary) and filtration, were directed to the RO module. The performance of RO is presented in Figs. 2 and 3 separately for OLD and NEW leachate. The recovery rate for both analyzed leachate was set on 60%, that is, 60% permeate and 40% concentrate. The assumed recovery rate was reached on average after 5.4 h for OLD leachate and 7.0 h for NEW leachate (Figs. 2 and 3).

In this time the average flux for OLD and NEW leachate was 38.8 and 29.3 L/m^2 h, respectively (Fig. 2). As the recovery rate increased, the permeate flux decrease was observed for both types of leachate. The most intensive flux decrease was noted in the first 15% recovery rate for OLD leachate and the first 20% recovery rate for the NEW one. Along with the percentage recovery increase, the salts in feed became more concentrated. This slowed down the RO process, causing the permeate flux to decrease.

Table 1
Characteristic of young (NEW) and matured (OLD) leachate from analyzed landfill

Average ± standard deviation			
Parameter	NEW leachate	OLD leachate	Standard limits*
pH	6.5 ± 0.09	7.70 ± 0.14	6.5 ÷ 9.0
EC	5.37 ± 1.23	8.19 ± 2.1	–
BOD/COD	0.5 ± 0.01	0.01 ± 0.01	–
COD	4,211 ± 796	1,403 ± 40	125
$N–NH_4^+$	753.3 ± 127	732.7 ± 41.2	10
TP	17.11 ± 1.62	2.01 ± 0.38	3
SO_4^{2-}	215.7 ± 52.3	377.1 ± 149	500
Fe	2.73 ± 0.97	4.30 ± 0.75	10
Cl^-	2,100 ± 469	1,389 ± 689	1,000
TDS	8,208 ± 883	4,596 ± 535	–
TOC	9,227 ± 630	954.1 ± 321	30
Mg^{2+}	410.5 ± 60	386.5 ± 50	–
Ca^{2+}	480.1 ± 50	450.0 ± 24	–

All in mg/L apart EC (mS/cm) and pH.
* Standard limits according to Regulation of the Minister of Environment of 18 November 2014 on the conditions to be fulfilled for the introduction of wastewater into water or land, and on substances which are particularly harmful to the aquatic environment (Dz.U.2014.1800 from 2014.12.16).

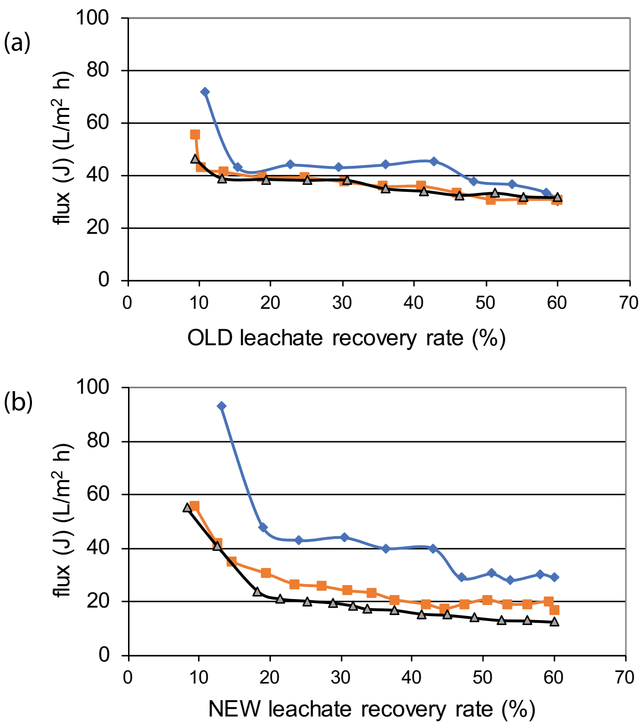

Fig. 2. OLD and NEW leachate flux (L/m² h) along with recovery rate (%).

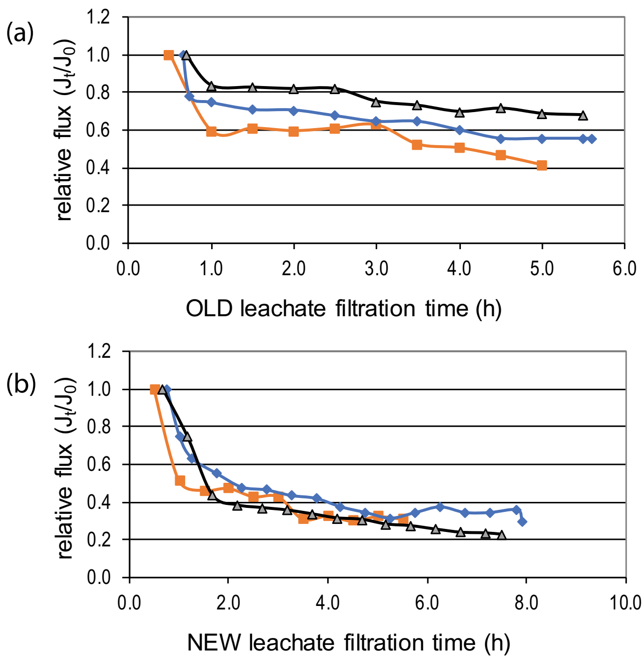

Fig. 3. Relative permeate flux (J_t/J_0) during filtration OLD and NEW leachate.

The maximum flux during high-pressure filtration of OLD leachate was 72 L/m² h. Relative flux, that is, the ratio of actual flux (J_t) to the initial one (J_0), decreased by over 50% at the end of the process. After the first hour of OLD leachate, filtration permeates flux oscillated within the range of 35 ÷ 45 L/m² h (i.e. 60% relative flux) and finally reached the value of about 30 L/m² h at the end of the filtration (Figs. 2 and 3).

The maximum permeate flux during the NEW leachate filtration process was 93.0 L/m² h. After the flux decline during the first 2 h of filtration, the permeate flux oscillated around 30 L/m² h, that is, 40% relative flux. This value dropped to 20 L/m² h at the end of the process, giving the relative flux of about 30%. It means that the filtration of young non-stabilized leachate with a higher content of organic matter influences the intensification of the fouling process, and decreases the efficiency of the RO process. This is also confirmed by the studies of Trebouet et al. [3], in which it is proved that the organic matter in the young leachate consists of low molecular mass particles, which facilitates their adsorption in the pores of the membrane causing their clogging.

The main reason for flux decrease, observed both during filtration of NEW and OLD leachates, was the fouling/scaling process and the increase in osmotic pressure caused by the high concentration (salinity) of the inlet (i.e. leachate and concentrate mixture) directed to the system. With high salinity of the feed, achievement a constant permeate flux requires application of higher pressure, which - in case of the process carried out at constant pressure and increasing feed concentration - caused a drop in the flux rate.

Chang et al. [37] explain that intensive blocking of membrane pores at the beginning of the filtration is caused by the intermediate blocking process, in which particles cumulate one on the other, most often causing reversible fouling, which can be removed by hydraulic washing. Further decrease of permeate flux took place in a gentler way, indicating the accumulation of dissolved organic and inorganic components of leachates as well as colloidal and suspended substances on the surface of the membrane. Decrease in the flow of young leachate was greater mainly because of more intensive fouling caused by organic matter. The concentration of TOC in young leachate was almost ten times higher than in matured leachate (9,227 and 954 mg/L, respectively for young and matured leachate) and the value of COD was almost five times higher than in old one (4,211 mg/L and 1,403 mg/L, respectively for young and matured leachate). The obtained result is in agreement with the study of Chianese et al. [34], who noted that reduction of permeate flux, observed upon increasing the leachate concentration, is due to the presence of the organic compounds described by the COD and TOC parameters. Kabsch-Korbutowicz and Majewska-Nowak [38] report that filtration of solutions containing organic matter and inorganic cations like Ca^{2+} causes a decrease in membrane permeability. The formed coordination complexes of inorganic ions with the organic matter have lower solubility than free humic particles favoring the fouling phenomenon.

An increase in the pH of the leachate from 6.5 (after acidification with sulfuric acid) to 8.0 was observed during the RO process (Table 2). The increase of influent pH may increase the possibility of scaling on the membrane surface as a result of precipitation of $CaCO_3$ [39,40]. On the other hand, along with the increase in pH, the carboxyl groups of the membrane material tend to deprotonate: -COOH → -COO-, limiting the phenomenon of fouling especially in the case of negatively charged molecules, e.g. natural organic matter (NOM) [39].

In the case of landfill leachates containing significant amounts of organic matter, the increase in pH during the filtration process may, therefore, be a beneficial effect. These observations are in agreement with the results obtained by Chang et al. [37], which indicate that during the RO process, landfill leachate with a life span of less than 3 ÷ 5 years should have a pH maintained at 7.5 ÷ 8.0, which limits fouling caused by the presence of organic matter in them. According to Peng [41], the suitable scope of pH of landfill leachate should be ranged from 7.15 to 8.15. Also, the results of Lee et al. [42] demonstrate that RO membrane fouling decreases with increasing pH. Nevertheless, an increase in pH causes a decrease in salt solubility and precipitation of sparingly soluble compounds, that is, carbonates, sulfates. It may lead to an intensification of the scaling, especially in the case of stabilized leachate containing a higher amount of mineral substances than the young leachate.

The permeate obtained from the RO process was clear and transparent. The removal rate for EC was 99.2% in OLD leachate and 98.2% in NEW leachate. The efficiency

Table 2
Analysis of the difference in the efficiency of reverse osmosis in treatment OLD and NEW leachate (*t*-Student test)

Parameter	Average removal rate (%)		*t*	*df*	*p*
	NEW leachate	OLD leachate			
pH	pH increase from 6.5 to 8.0	pH increase from 6.5 to 8.0	–	–	–
EC	98.9	99.2	0.650	4	0.55
COD	94.7	92.1	0.551	4	0.91
$N–NH_4^+$	94.4	97.1	0.583	4	0.59
TP	98.5	99.3	–1.219	4	0.29
SO_4^{2-}	88.6	83.2	1.011	4	0.37
Fe	99.7	99.7	1.044	4	0.36
Cl^-	99.4	99.7	1.189	4	0.30
TDS	99.4	96.0	1.096	4	0.33
TOC	94.1	92.3	–1.308	4	0.26
Mg^{2+}	98.9	99.2	0.568	2	0.63
Ca^{2+}	92.8	94.0	0.544	2	0.64

t - result of *t*-Student test, *df* - degrees of freedom, *p* - probability level in organic compounds retention for NEW leachate and was 94.7% for COD and 94.1% for TOC. These substances were removed from OLD leachate with efficiency of 92.1% and 92.3%, respectively. The efficiency of RO in removing of N–NH_4^+ was 97.1% for OLD and 94.4% for NEW leachate, TP - 99.3% for OLD and 98.6% for NEW leachate, and calcium - 94.0% for OLD and 92.8% for NEW one. Almost the same removal rate was obtained for iron (97%), chloride (99.4 ÷ 99.7%), manganese (98.9 ÷ 99.2%). The lowest removal rate - 88.6 % and 83.2%, respectively for young and matured leachate - was observed for sulfates. This is due to the introduction of additional sulfates into the RO system in the form of sulfuric acid used to acidify the leachate before the RO process. The sulfate ions together with the Ca^{2+} cations form sparingly soluble calcium sulfate, which can precipitate from the solution intensifying the scaling. The efficiency of RO for the rest of the analyzed parameters was over 90%. The performed *t*-Student test showed that observed differences are not statistically significant (Table 2).

4. Conclusions

Young and matured leachate were directed to the RO module for analysis of performance and efficiency of RO in treating landfill leachate.

- The average permeate flux, obtained during the RO process, was higher for OLD leachate. The relative flux in this process was about 40% at the end of filtration, while in the case of NEW leachate it dropped to about 20%.
- The higher permeate flux during filtration of OLD leachate resulted in a shorter time of achieving the assumed recovery rate.
- The main reason for the flux decline of NEW leachate was a higher content of organic matter, which caused the intensification of the fouling process.
- During the filtration of leachate, their pH increased from 6.5 to 8.0. In the case of matured leachates, this may affect the scaling intensification as a result of decrease of salt solubility and precipitation of hard soluble compounds.
- The efficiency of purification of young and stabilized leachate by RO was similar, but taking into account the operation time of the RO system, the final treatment efficiency was achieved faster in the case of filtration of stabilized leachate.

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