TREATMENT OF LANDFILL LEACHATE BY MEANS OF PRESSURE DRIVEN MEMBRANE OPERATIONS

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Abstract

Since municipal landfill leachates are very often difficult to be biodegraded, integrated systems combining several unit processes are frequently applied to achieve a satisfactory level of purification. The paper discusses the results of investigations into the application of pressure membrane processes: ultrafiltration, nanofiltration and reverse osmosis to the treatment of landfill leachate. The investigations compared the effectiveness of the biological treatment with the effectiveness of two integrated systems: activated sludge method – ultrafiltration – reverse osmosis, and nanofiltration – reverse osmosis.

Streszczenie

Z uwagi na fakt, że odcieki ze starych składowisk odpadów komunalnych są trudno biodegradowalne, coraz częściej, aby osiągnąć odpowiedni stopień ich oczyszczenia, stosowane są układy zintegrowane będące połączeniem kilku procesów jednostkowych. W artykule omówiono wyniki badań oczyszczania odcieków z zastosowaniem ciśnieniowych technik membranowych, a mianowicie: ultrafiltracji, nanofiltracji i odwróconej osmozy. Porównano efektywność oczyszczania tych ścieków metodą biologiczną osadu czynnego z dwoma układami kojarzącymi metody osadu czynnego z ultrafiltracją i odwróconą osmozą oraz nanofiltrację z odwróconą osmozą.

Keywords: **Treatment of landfill leachate; Pressure-driven membrane techniques; Hybrid systems.**

1. INTRODUCTION

Comparative studies of various methods of municipal solid waste elimination (disposal in the ground, combustion, composting) carried out in many countries have shown that the disposal on the surface of the ground is the cheapest method. Apart from economical aspects, this method eliminates the unfavourable impact of the waste on the environment and allows to monitor degradation of wastes into relatively neutral substances.

In Poland, leachate is deposited either in cesspits and then transferred to municipal wastewater treatment plants (50% of landfill sites) or discharged into a sewage system (6% of landfill sites). 27% of sites posses internal wastewater recirculation. Other sites discharge leachate directly into environment i.e. surface water [1].

One of the most important problem concerning landfill site management deals with the intake and treatment of leachates. Its high load which differs in particular seasons of the year and its divergent composition make the treatment of such wastewater much more difficult in compare with municipal sewage [2-4]. Since landfill leachates contain considerable amounts of toxic organic and inorganic compounds, they cannot be discharged directly into sewers. Thus, they are usually drained and stored in a reservoir from which they are directed back to the landfill site or transported to local biological sewage treatment plants. But, since leachates are not directly biodegradable, they require other physical and chemical methods which support the biological ones. Therefore, studies on treatment of landfill leachates using pressure driven membrane operations have been carried out for the past few years [5-14]. Those methods unlike biological treatment do not neutralize the contaminants. They actually allow to separate the sewage stream into two, concentrated one (with high contaminants load), which is usually directed back to the landfill, and into purified water stream, which can be discharged into natural reservoirs or sewers.

The preliminary tests were focused on comparison of the effectiveness of biological treatment of leachate with the effectiveness of two integrated systems. The first one combined the biological process with ultrafiltration and reverse osmosis, whereas the latter comprised only of pressure driven membrane operations i.e. nanofiltration and reverse osmosis.

2. MATERIALS AND METHODS

2.1. Raw material

The treated leachate was generated in the landfill site "Lipówka" intended for the storage of municipal waste from Dąbrowa Górnicza (southern Poland). The investigations revealed its high stability in respect of both organoleptic characteristics as well as chemical composition. The ratio BOD_5/COD of all the taken samples was very low and ranged from 0.1 to 0.4, which indicated the resistance of the leachate to biodegradation and excluded the possibility of easy degradation of the contaminants by the activated sludge method. The correlation ratio between the amounts of ammonium and total nitrogen was also constant. Almost all total nitrogen was in form of ammonium and the contribution of the remaining nitrogen forms was negligible. Table 1 presents the contamination indexes which characterize the tested raw leachate.

The control of the leachate composition and effectiveness of its treatment covered standard parameters such as pH, COD, suspended solids and conductivity [15].

2.2. Methods

The first stage of the leachate treatment dealt with the biological method of activated sludge.

The process was carried out in an SBR phase reactor which was fed with the leachate periodically once a day. This technique, however, allowed to oxidize partially the organics present in the wastewater and remove nitrogen in organic and ammonium form.

30 dm^3/d of the wastewater was pumped by peristaltic pump from a raw leachate tank into the reactor with a peristaltic pump and after purification and sedimentation, into a purified leachate tank. The leachate was aerated with an SPP-30-GJ-2 pump (by Hiblow).

The biological reactor suitable for leachate treatment was fed with the activated sludge taken from a domestic sewage treatment plant. The adaptation of microorganisms lasted ca 2 months. The reactor operated in one cycle during both adaptation period and actual investigations. The duration of particular steps of the cycle was as follows:

– flow of the leachate in to the reactor 0.75 h
– aeration 21.0 h
– sedimentation 1.0 h
– discharge of the purified leachate from the reactor 1.0 h
– idle time 0.25 h.

In order to assess the physiological state of activated sludge biocenosis, its respiration activity was monitored during the tests. Therefore, samples of activated sludge were taken from the reactor and placed in an isolated chamber equipped with an oxygen probe where a reduction in oxygen concentration with time was measured. The linear decrease in oxygen concentration allowed to calculate the rate of oxygen consumption by microorganisms expressed in $mgO_2/dm^3 \cdot h$. Oxygen concentration in the aeration chamber during the process was kept at 2 mgO_2/dm^3.

Since the biological treatment of the leachate did not yield satisfactory results and the contamination indexes of the purified wastewater did not allow it to be discharged into receiving water. According to that the subsequent stage of the research focused on its post-treatment, firstly by ultrafiltration and then reverse osmosis. The initial testing of ultrafiltration membranes with the

Table 1.
Characteristics of landfill leachate from the municipal landfill site "Lipówka"

Loading index of landfill leachate	Unit	Mean value
pH	-	8.0
BOD_5	mgO_2/dm^3	331
COD	mgO_2/dm^3	1183
Ammonium nitrogen	mg NH_4^+-N/dm^3	743
Nitrite nitrogen	mg NO_2^--N/dm^3	trace values
Nitrate nitrogen	mg NO_3^--N/dm^3	0.8
Total solids	mg/dm^3	240
Suspended solids	mg/dm^3	21

post biological treatment leachate allowed to select a membrane which showed the highest efficiency in retaining activated sludge and high molecular weight substances. Ultrafiltration was conducted in the cross-flow mode on flat membranes from polyvinyl chloride (PVC) and tubular membranes from polysulfone (PSf) prepared in a laboratory. The process was carried out under a transmembrane pressure of 3×10^5 Pa, with crossflow velocity of 2.5 m/s and a temperature of 298K. The operating parameters of reverse osmosis were as follows: transmembrane pressure 2.76×10^6 Pa, flow rate 1.5 m/s and temperature 298K. In case of nanofiltration, the transmembrane pressure was 2.07×10^6 Pa. The membranes applied in high pressure processes were made of cellulose acetate by Osmonics.

During the studies the transport and separation characteristics of the ultrafiltration membranes were investigated. The transport properties were determined by calculation of a dependence of the volumetric deionized water flux on transmembrane pressure within the range of 0.5×10^5 Pa – 3×10^5 Pa (PVC) and 0.5×10^5 Pa – 2.5×10^5 Pa (PSf). The processes were carried out at the temperature of 298 K.

The separation properties of the ultrafiltration membranes were determined by testing them with a dextran solution of nominal molecular weight 200 000, obtaining 10% of the feed each time. A pressure of 3×10^5 Pa, cross-flow velocity of 2.5 m/s and temperature of 298 K were applied. The tests enabled to obtain the cut-off values for particular membranes.

Concentrations of the dextran in the feed and permeate were determined, and next retention coefficients were calculated from the below equation:

$$R = (1-C_p/C_f) \times 100\% \qquad (1)$$

where: C_p, C_f – concentration of the substance in the permeate and feed, mol/dm^3.

The distribution of dextran molecular weights was obtained by means of gel permeation chromatography applying a “Shimadzu” chromatograph. The concentration of the dextran in the permeates was calculated from GPC chromatogram, assuming that its concentration in the sample introduced to the chromatographic column is directly proportional to the area under the peak on the chromatogram.

The separation properties of the nanofiltration and osmotic membranes were not determined because they were given by the producer [16].

The final stage of the research focused on the effectiveness of leachate treatment in the second integrated system which comprised of pressure driven membrane operations i.e. nanofiltration and reverse osmosis. Both processes were carried out under the transmembrane pressures recommended by the membrane producer i.e. 2.07×10^6 Pa for nanofiltration and 2.76×10^6 Pa for reverse osmosis. The flow rate of the leachate over the membrane surface was 1.5 m/s in both processes, and the temperature was 298 K.

Prior to nanofiltration, the raw leachate was additionally filtered (filter 50μm) to remove suspended solids. Due to the elevated pH, the leachate was acidified with hydrochloric acid to a pH equal to 6.0. Otherwise, the excessive pH value of filtrated wastewater might have an impact on transport and separation properties of the membrane. At the same time it could cause the hydrolysis of acetate cellulose from which the membrane was made.

3. RESULTS AND DISCUSSION

3.1. Biological treatment of leachate on activated sludge

Fig. 1 illustrates the changes in COD of the raw leachate treated by activated sludge.

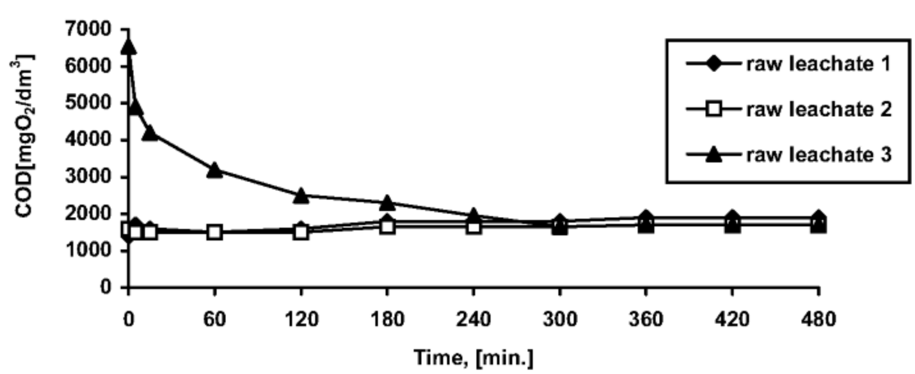


Figure 1.
Dependence of COD concentration of raw landfill leachate on the time of its biological treatment

Table 2.
The characteristic of raw landfill leachate and treated by the biological method of activated sludge

Loading index of landfill leachate	Unit	Raw landfill leachate	Landfill leachate after biological treatment
COD	mgO_2/dm^3	1600	1680
Suspension	mgO_2/dm^3	26	225
Dry matter	mgO_2/dm^3	164	395
pH		8.0	8.7
Conductivity	mS/m	9.5	11.2

It has been found that when COD of the raw leachate was low and did not exceed 2000 $mg/O_2/dm^3$ (raw leachate 1 and 2), the raw leachate contained large amounts of non-biodegradable compounds. Multiple attempts to grow biocenosis capable of degrading those refractive compounds resulted in a negligible removal of COD (<10%). A gradual decrease in respiration activity of activated sludge micro-organisms and an increase in dissolved oxygen concentration in the reactor were also found during the tests. No significant changes were observed in the sludge volume index.

Much different situation was found when the COD was high (raw leachate 3-6548 mgO_2/dm^3). In this case, aeration of the leachate with activated sludge caused a rapid degradation of organics. Five hours after the process start, 75% decrease of COD was observed and its obtained value was 1657 mgO_2/dm^3.

However, COD concentration in high-loaded wastewater could not be decreased below the COD level in low-loaded wastewater i.e. 1500-2000 mgO_2/dm^3. After this time, it was impossible to lower the index permanently. The characteristic of the wastewater after biological treatment are shown in Table 2.

The results of the tests showed a low susceptibility of the investigated wastewater to biodegradation. Therefore, it could not be discharged into ground waters without additional treatment.

3.2. Determination of transport and separation characteristics of applied membranes

3.2.1. Ultrafiltration and microfiltration membranes

Flat membranes made of polyvinyl chloride (PVC) and tubular membranes made of polysulfone (PSf) were used during the studies.

The PVC membranes were formed by phase separation. The thickness of the cast film, which was a mixture of polymer in dimethyl formamide, was 0.2×10^{-3} m and the active surface of membranes was 0.0155 m^2. The

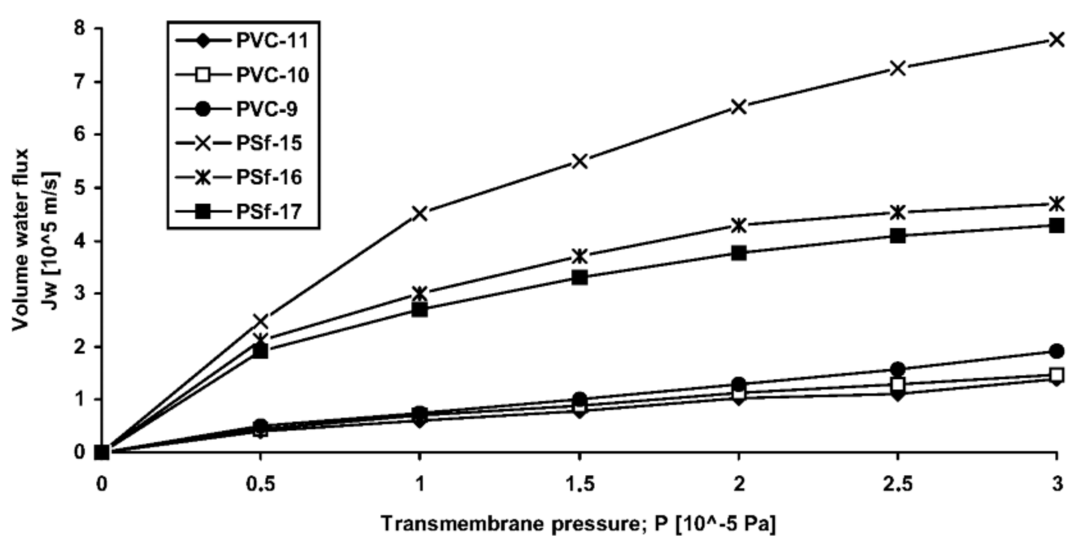


Figure 2.
Dependence of volume water flux on transmembrane pressure for PVC and PSf membranes of different polymer concentrations in the casting solution

membranes were of different compactness and thus different transport and separation properties due to the changes in polymer concentration in the casting solution during their preparation. Three membranes of 9, 10 and 11% wt. polymer content were examined. They were designated as PVC-9, PVC-10 and PVC-11.

The tubular polysulfone ultrafiltration membranes were also prepared by phase separation from the casting solution which was a mixture of polysulfone in dimethyl formamide. The thickness was 0.25×10^3 m and the active surface was 0.025 m^2. Polymer concentrations in the casting solution were 15%wt, (PSf-15), 16%wt. (PSf-16) and 17%wt. (PSf).

The transport characteristics of the membranes were described by the dependence of the volumetric water flux on transmembrane pressure illustrated in Fig. 2.

The efficiency of the membranes depended on polymer type and its concentration in the casting solution. The tubular polysulfone membranes showed several times higher water flux than the PVC membranes. The difference in the volumetric water flux between PSf-15 and PSf-17 was 42.3% for the transmembrane pressure of 2×10^5 Pa.

The separation properties of the membranes were determined applying a dextran solution of 5 g/dm^3 of nominal molecular weight 200 000 Da (produced by Polfa S.A., Kutno, Poland) to which sodium azide, which acted as bacteriostatic agent, was added. The concentration of sodium azide was 200 mg/dm^3.

Fig. 3 shows the relationships determined for PVC and PSf membranes, in which the retention coefficient on the abscissa corresponds to the molecular weight range.

The obtained results suggest that the PVC membranes are ultrafiltration membranes characterized by the following cut-off values:

- PVC-9 – 55 kDa
- PVC-10 – 30 kDa
- PVC-11 – 20 kDa.

The PSf membranes may be classified as microfiltration membranes due to low retention coefficients of the dextran which molecular weight was 200 kDa (<20%).

Fig. 4 and Fig. 5 depict the permeate fluxes obtained during ultrafiltration of the post biological treatment leachate.

The specific membranes exhibited various degrees of permeability. The flux in case of PSf-15 was highest and this was the only membrane which retained suspended matter completely. In the case of the flat PVC membranes, they all removed suspended matter. However, the permeate flux obtained as a result of leachate filtration on the most open membrane (PVC-9) was four times lower than on PSf-15. Therefore, the selection of this membrane for further tests was based on the value of the volume permeate flux.

3.2.2. Nanofiltration and osmotic membranes

The research employed nanofiltration (SF) and osmotic (SS) membranes whose characteristics were described by the manufacturer are given in Table 3.

The dependence of the volumetric water flux on

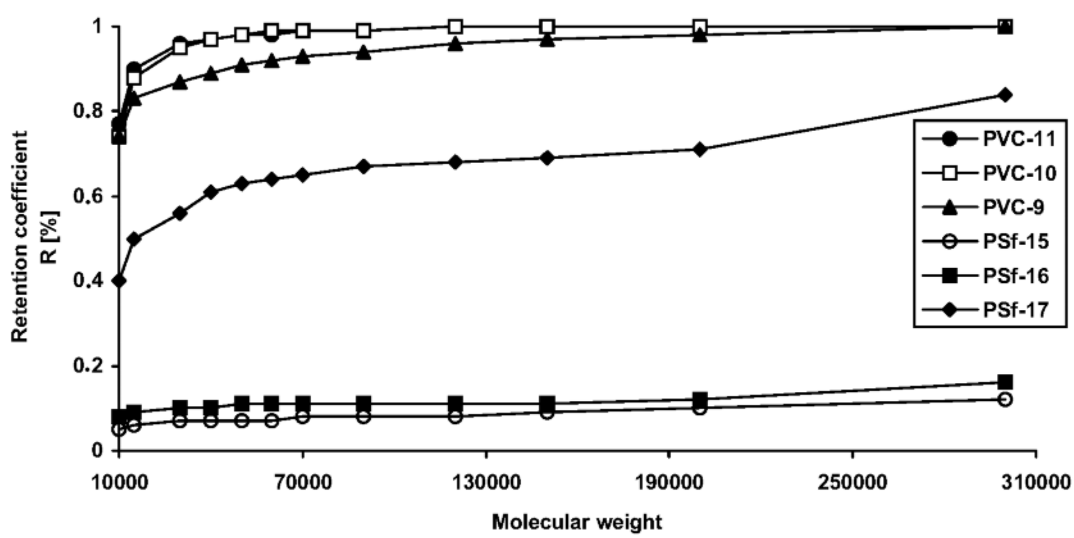


Figure 3.
“Cut-off” curves obtained from gel permeation analysis of feeds and permeates for PVC and PSf membranes

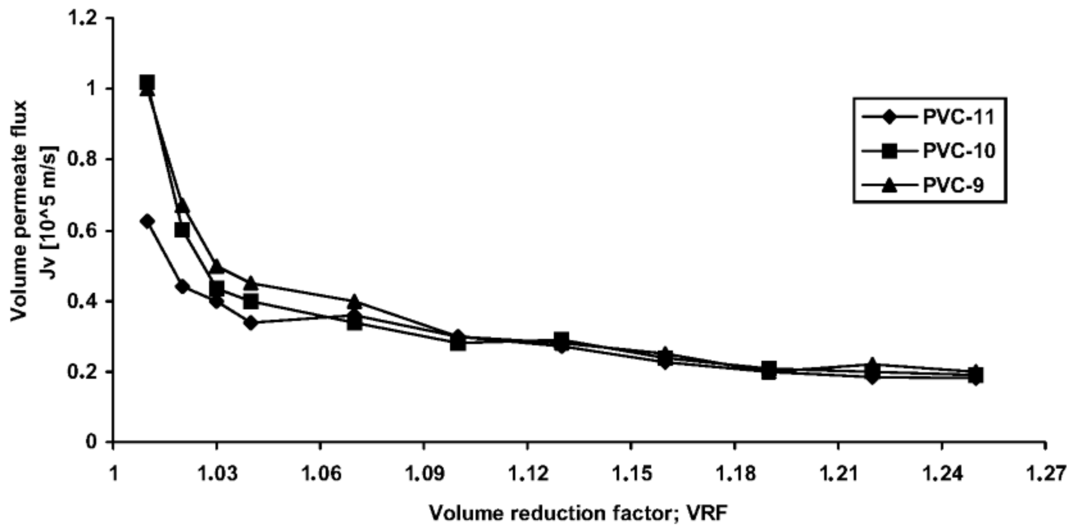


Figure 4.
Dependence of volume permeate flux on volume reduction factor for flat PVC membranes (ΔP = 0.3 MPa, u = 1.5 m/s, T = 298 K)

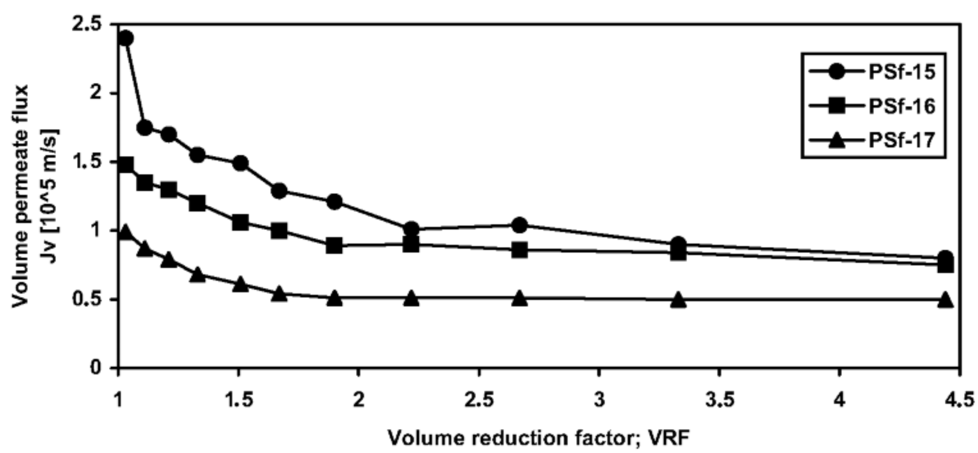


Figure 5.
Dependence of volume permeate flux on volume reduction factor for tubular PSf membranes (ΔP = 0.3 MPa, u = 2.5 m/s, T = 298 K)

Table 3.
Characteristics of the SEPA CF nanofiltration membrane (SF) and osmotic membrane (SS) [16]

Membrane	Retention coefficient NaCl, [%]	Transmembrane pressure, $\times[10^{-6}$Pa]		pH	Maximum temperature, [^{0}C]	Permissible concentration of available Cl; mg/dm^3
		Recommended	Maximum			
NF - SF	85	2.07	2.07	2-8	50	2
RO - SS	98	2.76	2.76	2-8	50	2

transmembrane pressure is given in Fig. 6.

For both membranes, the correlations were of linear character and were described by the following equations:

$$J_{W(SF)} = 5.80\times10^{-12}\,\Delta P + 8{,}00\times10^{-7} \qquad (2)$$

$$J_{W(SS)} = 1.80\times10^{-12}\,\Delta P + 5{,}50\times10^{-18} \qquad (3)$$

For the pressure of 3×10^6 Pa, the volumetric permeate flux for the SF membrane was 4.5 times higher than the flux for the SS membrane. It was associated with a more compact structure of the SS membrane.

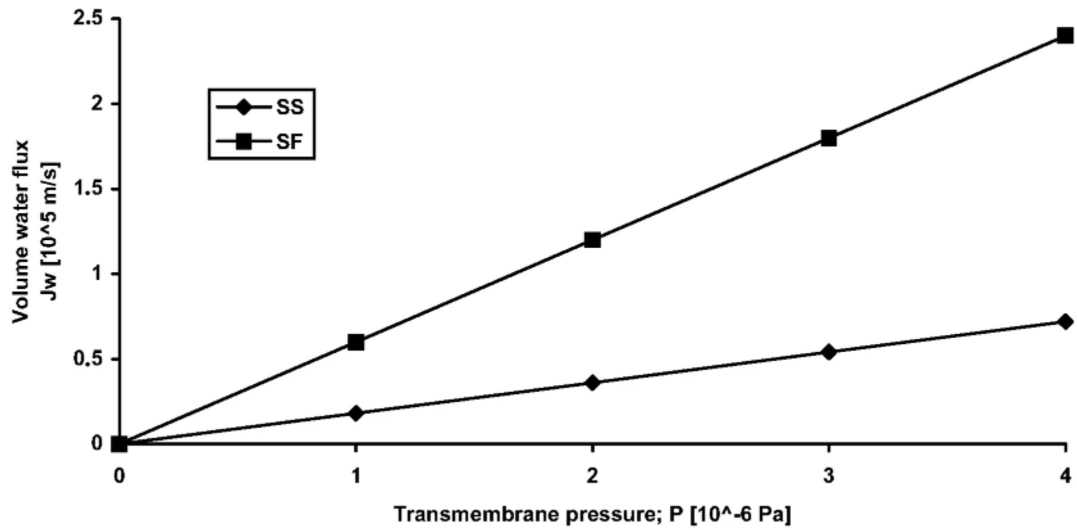


Figure 6.
Dependence of water flux on transmembrane pressure for nanofiltration SF membrane and osmotic SS membrane, (u = 1.5 m/s, T = 298 K)

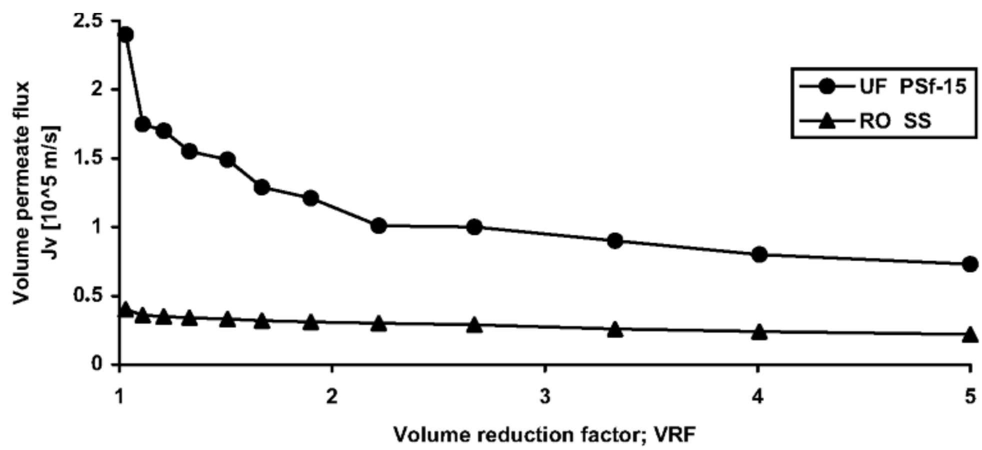


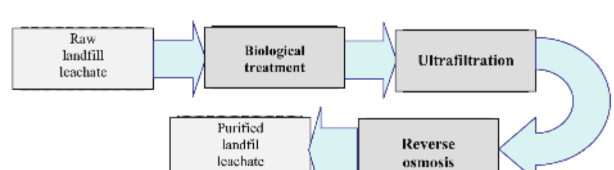
Figure 7.
Dependence of permeate flux on volume reduction factor during ultrafiltration and reverse osmosis of landfill leachate purified by means of biological method ($\Delta P_{(UF)}$ = 0.2 MPa, $u_{(UF)}$ = 2.5 m/s, $\Delta P_{(RO)}$ = 2.07 MPa, $u_{(RO)}$ = 1.5 m/s, T = 298 K)

3.3. Treatment of leachate by the integrated system: activated sludge – ultrafiltration – reverse osmosis

Since the biological methods did not ensure a satisfactory level of purification, the leachate was additionally treated with ultrafiltration and reverse osmosis processes.

Ultrafiltration took place after the biological treatment and completely removed suspended solids and high molecular weight compounds from the leachate stream, protecting the osmotic membranes from fouling. The low molecular weight organic compounds and inorganic salts left in the wastewater were removed almost completely during reverse osmosis.

The scheme of the integrated system is illustrated below:



Ultrafiltration was carried out in the concentrating system. The obtained dependence of the permeate flux on volume reduction factor is illustrated in Fig. 7.

Table 4.
Results of physicochemical tests on landfill leachate treated in the system combining the processes of biological treatment, ultrafiltration and reverse osmosis

Loading index of landfill leachate	Unit	Raw landfill leachate	Landfill leachate after biological treatment	Landfill leachate after ultrafiltration	Landfill leachate after reverse osmosis
pH	-	8.0	8.6	8.8	7.6
COD	mgO_2/dm^3	1780	1660	846	56
Suspended solid	mg/dm^3	26.3	225	0	0
Conductivity	mS/m	8.8	10.0	10.0	1.2
Cl^-	mg/dm^3	1290	1180	1100	478
Ca^{2+}	mg/dm^3	0.09	-	0.03	0.01
Fe^{3+}	mg/dm^3	8.92	-	0.15	0.08
Zn^{2+}	mg/dm^3	3.17	-	0.26	0.21
Cd^{2+}	mg/dm^3	0.01	-	trace values	trace values
Cu^{2+}	mg/dm3	2.15	-	trace values	trace values

Table 5.
Results of physicochemical tests a landfill leachate treated in the system combining the processes of nanofiltration and reverse osmosis

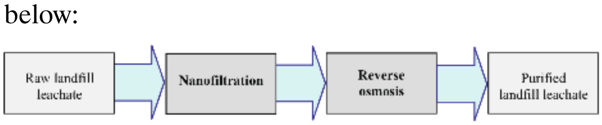
Loading index of landfill leachate	Unit	Raw landfill leachate	Landfill leachate after ultrafiltration	Landfill leachate after reverse osmosis
pH	-	8.0	8.4	7.4
COD	mgO_2/dm^3	1780	197	56
Suspended solid	mg/dm^3	26.3	0	0
Conductivity	mS/m	8.8	11.4	1.6
Cl^-	mg/dm^3	1290	1156	321
Ca^{2+}	mg/dm^3	0.09	trace values	trace values
Fe^{3+}	mg/dm^3	8.92	0.15	0.02
Zn^{2+}	mg/dm^3	3.17	0.26	0.01
Cd^{2+}	mg/dm^3	0.01	trace values	trace values
Cu^{2+}	mg/dm3	2.15	trace values	trace values

A fivefold reduction of the initial volume of the leachate in the process caused a threefold reduction of the volume permeate flux. The rapid decrease in filtration velocity, especially in the first phase of leachate concentration, was probably the effect of fouling.

The volumetric permeate flux obtained in the next stage of leachate treatment – reverse osmosis – decreased by 25% for VRF = 5. The treatment effects obtained in the discussed system are given in Table 4.

3.4. Integrated nanofiltration and reverse osmosis leachate treatment system

The scheme of the integrated system is presented below:



Prior to nanofiltration, the raw leachate was filtered to remove suspended solids. It was also neutralized to pH = 6.5.

Table 5 shows physicochemical parameters of the leachate treated in the discussed system.

The obtained results clearly show that the treatment of leachate by the integrated nanofiltration and reverse osmosis system ensures a satisfactory purification level. Nanofiltration removed multivalent metals to a large extent, but the leachate still contained considerable amounts of chlorides, and COD, despite marked reduction, remained at excessive levels. The subsequent application of reverse osmosis enabled a further reduction in COD and chloride ion concentration which allowed to discharge the purified leachate into natural water reservoir.

A comparison of the changes in the volume permeate fluxes in respect of the volume reduction factor for both processes is given in Fig. 8.

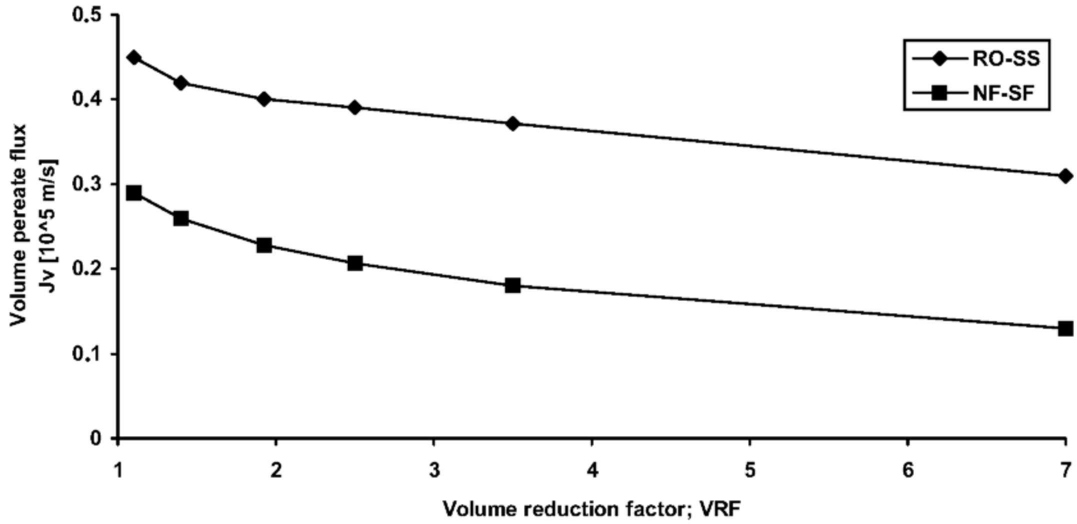


Figure 8.
Dependence of permeate flux on volume reduction factor in nanofiltration and reverse osmosis ($\Delta P_{(NF)}$ =2.07 MPa, $u_{(NF)}$ = 1.5 m/s, $\Delta P_{(RO)}$ = 2.76 MPa, $u_{(RO)}$ =1.5 m/s T = 298 K)

The volumetric permeate flux obtained during reverse osmosis is higher than the flux obtained in the same process but carried out after ultrafiltration. This can be explained by the fact that the leachate pre-treated by nanofiltration contains negligible amounts of solids which could cause membrane fouling or precipitation of sparingly soluble salts.

4. CONCLUSIONS

The investigations showed that the unit biological treatment of leachate with activated sludge did not ensure a satisfactory purification degree enabling the leachate to be discharged into receiving water.

Although the introduction of the next stage i.e. ultrafiltration allowed to remove suspended solids completely and partially removed high molecular weight compounds, COD of the treated leachate still significantly exceeded permissible values. The desired effect was achieved after application of reverse osmosis.

The results also revealed that the purified leachate could not be discharged into receiving water after it was treated only by nanofiltration. Further treatment by reverse osmosis was required.

Both integrated systems tested in the studies ensured a satisfactory purification level of municipal waste and enabled it to be discharged into natural water reservoirs.

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