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Performance of VSEP vibratory membrane filtration system during the treatment of landfill leachates

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Abstract

In this study the performance of a vibratory shear enhanced unit (VSEP) for the treatment of raw stabilized leachate produced during landfill of municipal wastes was assessed. For this purpose, four different membrane types were examined for the treatment of leachates, i.e., one for microfiltration (with mean pore diameter 0.1 μm), two for ultrafiltration (with molecular cut-off 100 kDa and 10 kDa) and one for nanofiltration (50% rejection of NaCl). The vibration amplitude of the membrane module was set to 25.4 mm. The treatment capability of the system was evaluated especially regarding the removal of organic load (COD), of suspended solids, of conductivity (dissolved solids), of N–NH^{4+} and of HA. The removal of organic matter in terms of COD value was exceeded 60% for all cases. Similar pattern of retention capacity was also observed for the case of suspended solids, regarding the respective turbidity values. Finally, the rejection of humic-like substances was highly depended upon the applied membrane process, as the lowest removal capacity was observed for microfiltration (about 47%), whereas the highest for nanofiltration (about 97%).

Keywords: Membrane filtration; Landfill leachates; Vibratory shear enhanced process; VSEP

1. Introduction

Leachates from municipal waste landfill sites are considered complex, heavily polluted and hazardous wastewaters [1,2]. Leachates composition is site and seasonal specific, since many factors (such as the origin of wastes, hydrogeological conditions in landfill and age of landfill) strongly affect the final concentration of contaminants. Among contaminants, organic loading and some inorganic salts, such as sodium chloride and carbonates, are present in considerable concentrations. An important group of organic matter in sanitary landfill leachates consists of humic-type substances, which are comparable with the humic substances identified in aquatic environments [3,4]. In general, humic substances, like humic and fulvic acids, are heterogeneous organic constituents

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of moderate molecular weight that contain both aliphatic and aromatic groups, which are considered non-biodegradable during biological wastewater treatment [5].

Due to the complex and specific nature of municipal landfill leachates, the selection of an appropriate treatment method depends mainly upon the nature of the waste stream to be treated [6]. In order to overcome these specific problems, leachates treatment processes include usually a combination of different techniques. However, today new treatment techniques that were initially designed for the purification of drinking waters and wastewaters are examined. Among these processes, membrane filtration and in particular reverse osmosis receive more attention, due to high treatment (removal) degrees of pollutants that can be achieved [7,8]. Moreover, in many studies is reported that reverse osmosis permeate meets the disposal limits (according to local legislation) of organic loading and ammonia [7–9]. However, several drawbacks have limited the use of reverse osmosis technique in large-scale applications; it is rather expensive due to pre- and post-treatment costs [3], often fouling phenomena deteriorate the systems performance and dissolved salts (especially chloride) concentration in leachates may rise too high resulting in high osmotic pressures and thus, in low treatability of leachates [10]. Nanofiltration process exhibits treatment characteristics between ultrafiltration and reverse osmosis and is less effective in the removal of both organic and inorganic contaminants. Trebouet et al. [3] studied the effect of landfill leachate pretreatment on nanofiltration performance and concluded that pH modification, prefiltration and coagulation with $FeCl_3$ did not enhance the retention ability or permeation fluxes. Yet, there is lack of sufficient available data, concerning the efficiency of membrane processes, other than reverse osmosis on the treatment of leachates.

The membrane filtration performance deteriorates due to concentration polarization and irreversible fouling caused by the formation of a boundary layer near the membrane surface and the accumulation of constituents on the membrane surface (i.e., formation of a cake layer), respectively. As a result, the operating costs of the membrane systems increase and the processes become less viable economically. There are many factors affecting membrane fouling, including membrane characteristics (molecular weight cut-off, MWCO, surface charge, hydrophobic properties and surface morphology), feed water composition and properties [pH, ionic strength, natural organic matter (NOM) presence, pollutant type and concentration] and membrane operation characteristics (pressure, temperature, cleaning) [11,12]. Among these factors, NOM is considered as a major foulant in membrane processes, as the interactions between NOM and the surface of membrane can cause severe flux decline.

In order to confront the concentration polarization and membrane fouling many techniques have been developed, based on the increase of the shear rate near the membrane surface. In crossflow filtration the process fluid passes tangentially to the surface of membrane, thereby the retention of the separated constituents onto the membrane surface is prevented. However, in most cases high fluid crossflow velocities are required in order to achieve adequate shear rate, especially when the process fluid contains high solids loading. These constrains enforced the development of advanced operating techniques, such as automatic backwashing, use of turbulence promoters or rotating disk modules [13,14].

Vibration shear enhanced filtration system (VSEP) is an alternative approach on dynamic filtration and has gained attention due to the improved operational characteristics, such as better control of membrane fouling phenomena. The pressure vessel moves in a vigorous vibratory motion tangent to the face of membranes, creating shear waves that prevent efficiently membrane fouling. VSEP filtration has been applied during several processes, such as the purification of pulp and mill paper circulation water [15], the treatment of yeast dispersions and bovine albumin solutions [16] and of dairy process waters [17], the separation of casein micelles from skimmed milk [18] and lately the treatment of leachates [8]. As it was reported by Takata et al. [19], the VSEP filtration module was effective for the prevention of fouling, when used for the treatment of river water. On the other hand, Bian et al. [20] has examined the effect of shear rate during concentration polarization of microfiltration and nanofiltration membranes, due to humic substances deposition on the membrane surface and concluded that the operation at maximum vibratory amplitude resulted at high removal efficiency during nanofiltration, even when the recovery rate reached 96%. Moreover, in a previous study Petala and Zouboulis [21] studied the removal efficiency of humic substances in the presence of suspended solids and found that VSEP system presented stable hydrodynamic behavior even when treating water with high concentrations of humic substances and inorganic clays. Chan et al. [8] investigated the removal of some organic and inorganic contaminants of leachates by reverse osmosis using VSEP system and concluded that substantial COD and ammonium nitrogen removals could be achieved. However, the hydrodynamic behavior and treatment ability of VSEP filtration system during the treatment of leachates has not been examined sufficiently.

The objective of this study was to evaluate the comparable performance of VSEP unit using different membranes (microfiltration, ultrafiltration and nanofiltration) for the removal of contaminants contained in stabilized municipal landfill leachates.

2. Materials and methods

2.1. Examined wastewater

The examined landfill leachates were obtained from Thessaloniki's municipal landfill. The considered municipal landfill, which was established in 1982, is located about 30 km from Thessaloniki (N. Greece) and serves more than 1,000,000 inhabitants. Stabilized (aged) leachate samples from a retention pond, located in the lower part of the landfill, of total volume 40 L were collected in 10 L plastic carboys and stored at 4°C for the experimental performance.

2.2. Tested membranes

Six different membranes were examined, one of them made of Teflon for microfiltration studies, two made of regenerated cellulose for ultrafiltration and one of polyamide for nanofiltration experiments. The used membranes were flat-disk modules with an effective membrane area of about 0.05 m^2. An additional drainage cloth that included polycarbonate track etched and polypropylene film was also used for membrane support. The membrane characteristics, as indicated by the manufacturer, are summarized in Table 1. The pure water flux (PWF) was determined prior to each experiment, in order to evaluate better the performance of each membrane.

2.3. Analytical methods

The flowrate was recorded at certain time intervals during each experiment, by simply using a calibrated volumetric cylinder and a timer. Furthermore, a thermocouple was introduced into the feed tank and the temperature was monitored throughout the tests.

The pH value of the feed and of the filtrates was measured using a pH meter 8519 model (Hanna Instruments). The concentration of humic materials in each sample was estimated spectrophotometrically using a Shimadzu spectrophotometer (Shimadzu Instruments) with the absorbance recorded at the wavelength 254 nm. All samples were measured without pH adjustment, introducing the sample into quartz cells, against deionized water, used as reference. The suspended solids concentration in each sample

Table 1
Overview of selected membrane properties, as indicated by the manufacturer of VSEP filtration system (New Logic International, USA)

Membrane code	T 0.1	C100	C10	TFC-50
Process	MF	UF	UF	NF
Material	PTFE (Teflon)	Regenerated cellulose	Regenerated cellulose	Polyamide/polysulfone
Cut-off diameter	0.1 µm	100 kDa	10 kDa	50% rej. NaCl
Maximum pressure (bar)	7	17	17	29
pH range (20°C)	1–12	1–11	2–11	3–11
Pure water flux (PWF) at 10 bar ($L\ m^{-2}\ h^{-1}$)	600*	600	100	80

*Operation pressure: 5 bar (1 bar = 10 197 $kg\ m^{-3}$).

was evaluated using the nephelometric turbidimeter Turbidity Lab-vis meter (Aqualytic). The electrical conductivity of leachate samples before, during and after membrane filtration was also measured at room temperature (about 20°C) using the HI 8733 conductivity meter (Hanna Instruments). Ammonium nitrogen and chemical oxygen demand were determined with Aqualytic spectrophotometer, according to respective standard tests [22].

The removal efficiency of a constituent (pollutant) by a membrane is defined as

$$R = \frac{C_o - C_p}{C_o} \tag{1}$$

where R is the removal efficiency of the membrane for a given pollutant at a defined hydrostatic pressure and feed solution concentration, whereas C_p and C_o are the concentrations of rejected components in the permeate and in the feed, respectively.

2.4. VSEP module

The vibratory shear enhanced filter (VSEP) series L was a semi-pilot scale module, manufactured by New Logic International (USA). The filter pack assembly consisted of two steel pressure plates and the polypropylene clamshell, which was the housing containing the installation of membrane. The filtration module consisted of an annular membrane of about 503 cm^2 area, with 13.5 cm outer radius (R_1) and 4.7 cm inner radius (R_2), in a circular housing placed at the top of a 1.5 m vertical shaft (Fig. 1). This shaft was mounted on a seismic mass and acted as a torsion spring, which transmitted the oscillations created by an eccentric drive motor. The membrane oscillated azimuthally in its own plane with an amplitude, dependent upon the frequency of drive motor. The shear rate that is created at the membrane is produced by the inertia of the fluid as in the case of Stokes layer near an oscillating plate [16]. The frequency of the oscillations was adjusted by an electronic controller with 0.01 Hz accuracy, while the resulting amplitude was recorded according to the pattern of appropriate black indicator marks, placed on the front of the clamshell. On the top of the spring was the permeate tubing that removed the permeate created by the membrane filtration action at atmospheric pressure. The concentrated stream was returned through the ‘process out’ line, as it is shown in Fig. 1. The return flow was passing through the flow limiter and the control valve, which allowed the fine adjustment of the outlet pressure. Inlet and outlet pressures were measured by Validyne

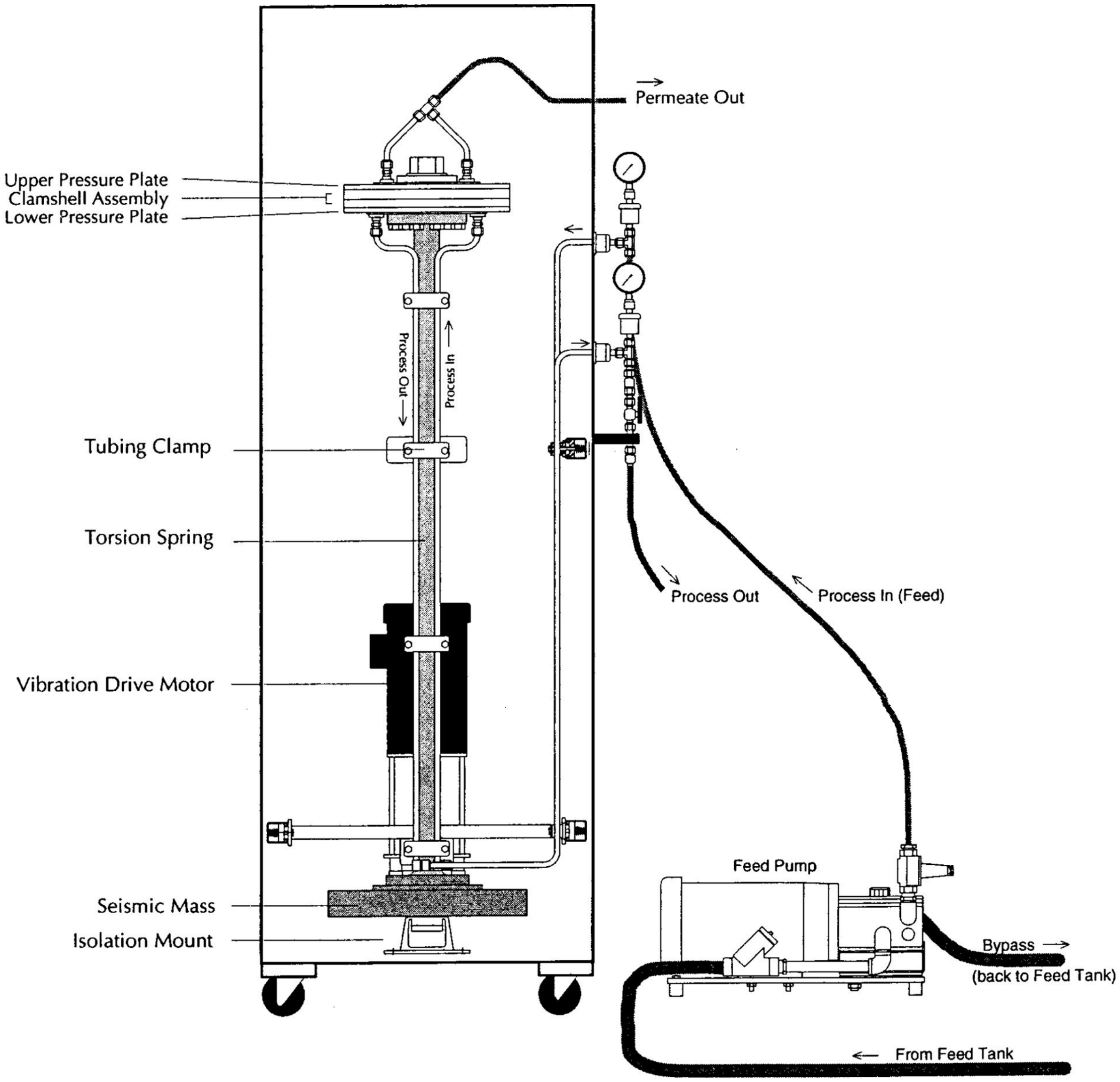

Fig. 1. Front view of the VSEP filtration system.

analog gauges, in order to determine the transmembrane pressure (TMP) as the mean of inlet and outlet pressures, since permeate was collected at atmospheric pressure. The manufacturer proposed (for safety reasons) that the outlet pressure should be at least 2 bar, when starting vibration, while the membrane should operate at pressures higher than 4 bar for oscillation amplitude 6.35 mm and higher than 8 bar for oscillation amplitude 12.7 mm [23].

During VSEP process, the maximum ($\gamma_{\max}$) and mean (γ_{w}) induced shear rate at the membrane surface was calculated by the following equations [16]:

$$\gamma_{\mathrm{w,max}} = 2^{1/2} d(\pi F)^{3/2} \nu^{-1/2} \quad (2)$$

$$\bar{\gamma}_{\mathrm{w}} = \frac{2^{3/2}(R_2^3 - R_1^3)}{3\pi R_2(R_2^2 - R_1^2)}\gamma_{\mathrm{w,max}} \tag{3}$$

where d is peak to peak vibration amplitude at the periphery of membrane (m), F is the vibration frequency (Hz) and ν is the kinematic viscosity of the fluid ($\mathrm{m^2\ s^{-1}}$).

3. Results and discussion

3.1. Development of shear rate and comparison with other filtration systems

The ability of VSEP system to induce increased shear on the membrane surface was initially examined. Fig. 2 shows the calculated values, according to Eqs. (2) and (3), for maximum and mean shear rate produced at the membrane surface during VSEP filtration of tap water at 20°C. The studied vibration frequencies and the corresponded amplitudes refer to the examined VSEP equipment. As it is shown, a relatively low increase in frequency settings resulted to an increase of vibration amplitude and hence to the produced shear rate on the membrane surface. In particular, as the amplitude increased from 6.35 to 24.5 mm, the corresponded maximum shear rate increased from about 19,500 to 78,000 $\mathrm{s^{-1}}$, while the mean shear rate at membrane surface rose from about 6,400 to 25,500 $\mathrm{s^{-1}}$. Moreover, in the case of an industrial VSEP machine the membrane elements are arrayed as parallel disks separated by gaskets. This disk stack is moved in a torsional oscillation, thus producing even higher shear rates of about 150,000 $\mathrm{s^{-1}}$ [23]. It is worth noting that for crossflow filtration systems, a crossflow velocity equal to 1 $\mathrm{ms^{-1}}$ produces shear rate around 1,500 $\mathrm{s^{-1}}$ [20], therefore the induced shear rate at the examined vibratory system was at least 3 times greater than in the conventional crossflow systems. In addition, a raise of crossflow velocity up to 5 $\mathrm{m\ s^{-1}}$ did not induce a maximum shear rate more than 25,000 $\mathrm{s^{-1}}$. Besides, it is important to stress out that in crossflow systems the shear originates in the process fluid, while in VSEP system the shear originates at the membrane surface. Therefore, crossflow systems depend significantly on the shearing forces of the feed slurry and high operating costs are required for the development of high shear rates during the filtration of viscous slurries.

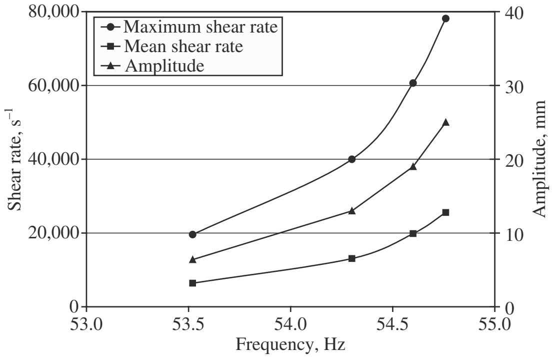

Fig. 2. Shear rate at the membrane surface for tap water at 20°C and vibration amplitude in relation to the applied frequency.

The development of shear in a rotating disk filtration system was examined by Jaffrin et al. [24], who calculated the maximum shear rate according to Eq. (4):

$$\gamma_{\mathrm{m}} = 0.0296\nu^{-0.8}(k\omega)^{1.8}\,R_d^{\,1.6} \tag{4}$$

where γ_{m} is the maximum shear rate that occurs at the disk rim and is obtained by setting $R_d = 7.25$ cm in (4), ω is the disk angular velocity, $k\omega$, the angular velocity of inviscid fluid in the gap and k is the velocity factor [24]. Taking into account that k is 0.42 for a smooth disk, the maximum shear rate for tap water at 20°C will be about 24,600 $\mathrm{s^{-1}}$ for angular velocity of 1000 rpm, while an increase of velocity to 2000 rpm would result to shear rate of about 85,800 $\mathrm{s^{-1}}$, according to Eq. (4). Moreover, the use of rotating disk, which is equipped with radial vanes rotating at high speed near a flat circular membrane ($k = 0.89$), yields even

Table 2
Physicochemical parameters of the examined landfill leachates

Parameter	Landfill leachate
pH	8.3
Conductivity (μS cm^{-1})	16,000
COD (mg L^{-1})	4100
Ammonium nitrogen (mg L^{-1})	580
Turbidity (NTU)	65
Absorbance at 254 nm (cm^{-1})	30

higher shear rates [25]. According to Eq. (4) the induced shear rate for tap water at 20°C will be about 95,200 s^{-1} for angular velocity of 1000 rpm, while an increase of velocity to 2000 rpm would result to shear rate of about 330,000 s^{-1}. It appears that in most cases, VSEP system induces higher shear rates, than the rotating disk system; however, for special configurations of disks the induced shear rates may be higher for the latter system. Nevertheless, filtration under high shear rates conditions limit fouling phenomena, thus improving membranes performance and making the water or wastewater treatment application of membrane filtration techniques more attractive.

3.2. Leachates treatment by VSEP filtration system

The examination of VSEP efficiency for the removal of organic matter (as presented by the conveniently measured parameters COD and ultraviolet absorbance at the wavelength of 254 nm) from landfill leachates included the investigation of the efficiency of various membranes. The main physicochemical properties of leachates are presented in Table 2. Comparing the parameter values with those obtained during a previous study [1], it was concluded that leachates were already stabilized (at least partially).

For the treatment of leachates four membranes were examined, as described earlier (see also Table 3). The experiments were conducted in semi-batch mode (concentration mode), i.e., the permeate was removed whereas the retentate was recirculated in the feed tank; while the TMP was maintained stable and the permeate flowrate was recorded at regular intervals. Vibration amplitude was set to 25.4 mm, according to stroboscopic indicators.

The final leachate volume after each filtration (semi-batch) process was equal to 0.5 L, resulting to volume recovery of 95% for microfiltration and ultrafiltration (100 kDa), and of 90% for ultrafiltration (10 kDa) and nanofiltration (since the initial feed volume was 5 L). The corresponding time to achieve the final concentration is also shown in Table 3. The system could reach even higher recovery rates, if the initial volume of leachates was higher. As shown in Fig. 3, it was observed that the permeate fluxes were stabilized after a certain period of time, although the system was operated in semi-batch (concentration)

Table 3
Landfill leachate treatment experimental conditions

Membrane process	Initial sample volume (L)	Final concentrate volume (L)	TMP (bar)	Vibration amplitude (mm)	Volume recovery (%)	Treatment duration (min)
Microfiltration (0.1 μm)	10	0.5	5	25.4	95	65
Ultrafiltration (100 kDa)	10	0.5	10	25.4	95	80
Ultrafiltration (10 kDa)	5	0.5	17	25.4	90	60
Nanofiltration (50% rej. NaCl)	5	0.5	20.4	25.4	90	150

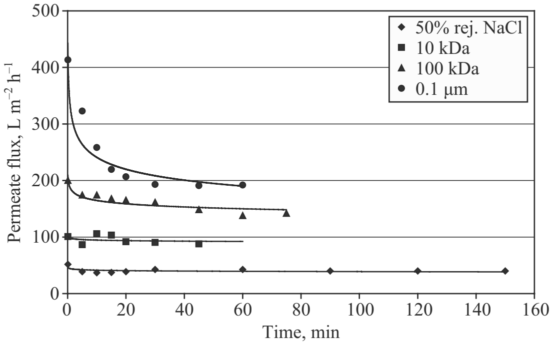

Fig. 3. Permeate flux with time during each membrane filtration process.

mode and the feed stream contained progressively higher concentrations of contaminants. Consequently, at the end of each process the permeate flux remained stabilized, while the retentate volume was about 0.5 L, indicating that the system could treat heavily contaminated wastewaters and control fouling efficiently.

The system reached stabilized flux conditions, after approximately 30 min filtration time during microfiltration and ultrafiltration with 100 kDa membrane, while during nanofiltration and ultrafiltration with 10 kDa steady-state flux conditions were achieved after just 15 min of filtration time. Petala and Zouboulis [21] reported a quite similar behavior during the treatment of various HA and inorganic clay dispersions. However, at this point it should be reported that feed temperature increased gradually during leachate treatment, unlike HA dispersion treatment, where feed temperature was quite stable. For example, feed temperature rose from 19°C to 30°C during leachate microfiltration, while during nanofiltration temperature increase from 20°C to 40°C. On the other hand, feed temperature during HA and clay dispersion varied between 20°C and 25°C. The increase of feed temperature, apparently affected the viscosity of leachates, thus it was considered that the stabilization of permeate flux was partially achieved due to temperature rise. The difference between initial and stabilized flux was substantially greater in the cases of microfiltration (from 400 to 192 L m^{-2} h^{-1}) and ultrafiltration with 100 kDa membrane (from 200 to 146 L m^{-2} h^{-1}), compared to ultrafiltration with 10 kDa membrane (from 100 to 90 L m^{-2} h^{-1}) and nanofiltration (from 50 to 40 L m^{-2} h^{-1}). This behavior was possibly attributed to phenomena, such as absorption of low molecular weight molecules inside the membrane pores and concentration polarization. However, in the case of membranes with smaller pore diameters adsorption is expected to take place to a lesser extend, while the produced shear prevented the further flux decline of permeate. It should be mentioned that it was not feasible to compare the leachate treatment results of this study with results of other investigators, since to our knowledge relevant data concerning micro-, ultra- and nano-filtration behavior with dynamic filtration system have been not published so far. Additionally, the majority of the studies that deal with the use of membranes for leachate treatment investigate the reverse osmosis application, while the examination of other membrane processes is limited [3,26].

The removal capabilities of contaminants of each membrane are depicted in Fig. 4. Conductivity reduction was proportional to the respective membrane pore diameter and it was observed

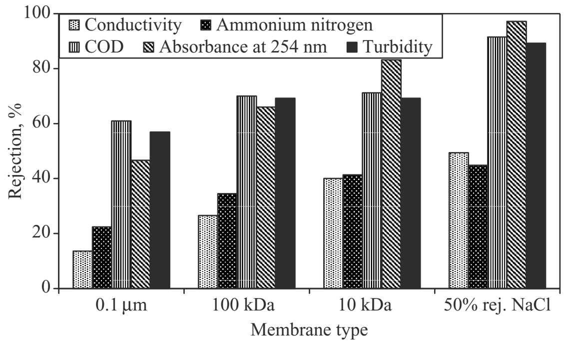

Fig. 4. Effect of different membrane filtration processes on the rejection (reduction) capacity of pollutants during the treatment of stabilized landfill leachates.

even after microfiltration (about 15%). Ultrafiltration resulted in a higher conductivity reduction that reached 30% in the case of 100 kDa and 40% in the case of 10 kDa membrane. The retention of was even greater by the application of 50% rej. NaCl membrane, since conductivity of the filtrate was about 8200 μS cm^{-1} in this case, corresponding to 49% reduction.

Ammonium nitrogen removal capacity was 22% and 45% for 0.1 μm and 50% rej. NaCl membranes, respectively. Organic matter removal, in terms of COD values, exceeded 60% in all cases. Furthermore, both ultrafiltration membranes presented similar COD removal capacities, about 70%, while microfiltration membrane retained 61% COD and nanofiltration was more efficient and reached up to 90%. However, it should be noted that even nanofiltration did not remove ammonium and COD below the discharge (legislation) limits; whereas Chan and his coworkers [8] found that VSEP using reverse osmosis membrane filter could achieve permeate concentrations of these parameters lower than the local discharge limits (200 mg/L for COD and 5 mg/L for ammonium nitrogen). The legislation limits for Thermaikos Gulf in Thessaloniki are 180 mg/L for COD and 35 mg/L for total ammonium content [27]. Similar pattern of retention capacity was also observed for suspended solids, as measured by turbidity. Turbidity removal was 57% for microfiltration, 69% for both ultrafiltration membranes and 89% for nanofiltration. Humic-like substances rejection depended upon the specific membrane process, thus the lowest removal capacity was observed for microfiltration, about 47%, while the highest for nanofiltration, about 97%. Certain differences between COD rejections and UV-absorbance reductions were recorded, probably because COD refers to refractory organic matter with lower molecular weights, as compared to that of humic substances [3]. Microfiltration yielded rather high rejections of COD and turbidity, when compared to ultrafiltration efficiencies. Although microfiltration is applied mainly for the rejection of suspended solids, it was found in this study that contaminants removal is also significant. The application of 100 and 10 kDa membranes during ultrafiltration experiments, showed that the 100 kDa membrane is more satisfactory, since the obtained removal capacities for both membranes were rather similar, whereas the 100 kDa membrane enabled higher permeate fluxes.

The removal of COD from leachates previously treated by coagulation–flocculation was examined by Silva et al. [28] who observed hardly 50% reduction after the sequential ultrafiltration experiments. Moreover, the highest removal was obtained after filtration with the 50 kDa membrane, whereas the 20 and 5 kDa membranes contributed to a lesser extend to COD removal. In this study, COD removal was much higher (about 70%) and was not dependent on the molecular weight cut-off of the membrane. Trebouet et al. [3], using a capillary nanofiltration membrane, found that the rejection capacity of ammonium nitrogen and COD were lower and did not exceed 21% and 80%, respectively, while the operation mode was different, since during the crossflow filtration both permeate and retentate returned to the feed tank. The lower contaminants removal could be partially attributed to the lower achievement of shear rate; Chianese et al. [7] found that COD rejection increased as the applied pressure increased, while Petala and Zouboulis [21] observed a slight increase of humic substances removal with increasing of the vibration amplitude (i.e., of the shear rate at the membrane surface). The increase of vibration amplitude induces higher shear rates on the surface of membrane, which corresponds to an increase of particle–particle collisions, thus to an increase of shear diffusion that forces the particles away from the membrane surface, i.e., back to the bulk solution [7]. Furthermore, the reduction of concentration polarization lowers the concentration of contaminants at the membrane surface and their diffusive transfer through the membrane, as well [21].

4. Conclusions

The performance of a VSEP unit by using microfiltration, ultrafiltration or nanofiltration membranes for the treatment of municipal landfill leachates was evaluated in this study. The hydrodynamic behavior of the system was satisfactory; the permeate flux remained stable (after a certain period of time), even when the feed stream contained progressively higher concentrations of contaminants, due to the recirculation of retentate to the feed tank. This was attributed to the ability of VSEP unit to develop high shear rates at the membrane surface by the effect of vibration. The shear rates were found to be much higher than those induced during crossflow filtration systems and comparable to those developed by the rotating disk filtration units. In addition, the induced shear rate enhanced the removal of contaminants, which was significantly higher, when compared with the achieved removal efficiencies obtained by other convenient membrane filtration systems. However, the examined processes could not remove the examined contaminants at levels below discharge limits, when applied alone. A combination of microfiltration or ultrafiltration with 100 kDa membrane and nanofiltration could be a promising alternative.

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