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TREATMENT AND UTILIZATION OF THE CONCENTRATE FROM MEMBRANE SEPARATION PROCESSES OF LANDFILL LEACHATES

Rafał NOWAK, Maria WŁODARCZYK – MAKUŁA¹ Czestochowa University of Technology, Department of Environmental Engineering Czestochowa, Poland

Abstract

The purpose of the paper was to assess the effectiveness of selected physico-chemical processes to improve the quality of retentates/concentrates obtained during the treatment of landfill leachates using membrane separation. Among the physico-chemical methods, Advanced Oxidation Process (AOP) and electrocoagulation were analysed. Landfill leachate resulting from the infiltration of waste mass by atmospheric precipitation as well as the dissolution and leaching of waste components are most often subjected to membrane separation. Permeate is usually discharged to the receiver, while the concentrate is recirculated and sprinkled on a waste pile. However, such action is only the retention of impurities in the body of the landfill and has an impact on the chemistry of raw leachates. Due to the very high concentrations of organic and inorganic compounds identified in the retentate, it is necessary to treat it, which will effectively reduce the amount of impurities in the leachate. Economic use seems to be another solution. An example would be growing energy crops but such application requires additional research.

Keywords: landfill leachate, retentate, membrane processes, electrocoagulation, AOP

¹Corresponding author: Czestochowa University o f Technology, 42 – 200 Czestochowa, 69 Dabrowskiego Str., +48 3250919, email: mwm@is.pcz.czest.pl

1. INTRODUCTION

As is known, landfill leachates are formed as a result of infiltration of rainwater through a layer of deposited waste and leaching and dissolution of waste components. The chemical composition of these leachates vary and depend on the type of waste and the duration of the landfill.

Landfill leachates are usually drained through a drainage system and collected in storage reservoirs called leachate reservoirs. In practice, they are often open earth reservoirs, but properly protected against leachate infiltration into the ground and penetration of components into the soil and water environment. From these averaging reservoirs, the leachate is usually directed to purification installations consisting of several unit processes. Subsequently, pre-treated and/or purified leachates can be discharged into a surface water receiver. Physical, chemical and biological processes are used in effluent treatment installations. Among the physicochemicals processes the following can be distinguished [17, 26, 27]:

- chemical neutralization and precipitation,
- coagulation with flocculation,
- filtration,
- ammonia stripping with air,
- pressure membrane processes.

The choice of processes depends on the qualitative composition of the leachate and the requirements of the water permit allowing the discharge of purified leachate to the environment. Pressure membrane processes such as ultrafiltration, nanofiltration and reverse osmosis (RO) are most widely used [1, 19, 20, 28, 38, 42]. Membrane modules are equipped with purification installations located at large landfills for many cities in Poland (e.g. Częstochowa). The basic process is RO reverse osmosis. The permeate can usually be drained to the receiver, which is allowed by the water law permit [14]. On the other hand, the retentate, constituting 20-25% of the leachate directed for treatment, is highly loaded with impurities retained as a result of the separation process [16, 35, 36]. In operated landfills, retentate is usually recirculated and spread on a pile of waste. This solution increases the concentration of impurities in raw leachate. Research by Tałałaj and Biedki showed that recirculation of the concentrate to the body of the landfill causes an increase in the concentration of COD, BOD and ammonium nitrogen, and changes in the leachate quality may affect the efficiency of raw leachate treatment in the RO installation [38, 39]. Thus, although recirculation of the concentrate to the landfill can accelerate the decomposition of waste components, dissolution and leaching, it does not comprehensively solve the problem. Drains from membrane cleaning are an additional waste water stream.

Due to the high concentration of organic and inorganic compounds, including toxic ones, further processing of the concentrate is necessary, consisting of improving the quality and/or recovery of useful ingredients. The aim of the study was to assess the effectiveness of selected physico-chemical methods that can be used to neutralize the concentrate obtained from membrane separation of landfill leachates and to identify the possibility of using the concentrate for economic purposes.

2. CHARACTERISTICS OF THE CONCENTRATE FROM MEMBRANE SEPARATION PROCESSES OF LANDFILL LEACHATES

In the literature, most of the works concern the characteristics of raw landfill leachates and permeate [2, 5, 10, 17, 33, 34, 37, 42], however, little information is available about the concentrated concentrate. Considering the large diversity of chemical compounds found in raw leachates and the high values of retention coefficients on membranes, it can be stated that most of the impurities will be retained in the concentrate. For example, removal of organic compounds is estimated to reach 99%. It should be emphasized that in the waste mixture there are continuous transformations of organic compounds that lead to the formation of macromolecular compounds, mainly humic substances. Studies have shown that the proportion of humic compounds in the total amount of soluble organic carbon can reach 60%. It is also observed that the molar masses of these compounds and the share of individual fractions vary (from several hundred to tens of thousands of Da), which depends on the age of the landfill. Chemical chemistry of landfill leachate has repeatedly confirmed that leachates also have toxic organic compounds such as: biologically active ingredients in plant protection products, polycyclic aromatic hydrocarbons (PAHs), halogenated organic derivatives or petroleum hydrocarbons (BTX) and benzenesulfonates (LAS), phthalates (DEHP) and many other connections [12, 16, 31]. Determination of individual compounds in leachates is random and usually the content of organic compounds is expressed in general COD and BOD indicators, and the proportion between these indicators informs about the biochemical transformations of these compounds in waste [16, 25]. RO installations also have high inorganic compound retention rates. An exemplary decrease in the value of inorganic compounds concentration recorded in the Tałałaj study was in the range of 83-97% [39]. Therefore, taking into account the high degree of separation both organic and inorganic compounds on membranes, they are present in high concentrations in concentrate. Exemplary values of concentrate quality indicators from the reverse osmosis process are presented in Table 1, while biogenic and other components - in Table 2.

Indicator	Unit	References			
		[9]	[15]	[30]	[32]
рН	-	8.2	-	7.2	7.5
Conductivity	mS/cm	31.3	-	51.4	27.8
COD	mg/dm ³	9700	26771	7450	2198
BOD ₅	mg/dm ³	3750	3750	365	-
Total organic carbon	mg/dm ³	-	-	1820	719
Ammonium nitrogen	mg/dm ³	3580	-	1580	2388
Total nitrogen	mg/dm3	-	-	4575	2571
Sulphur	mg/dm ³	2657	-	453	-
Chloride	mg/dm ³	3530	-	6430	1823
Iron	mg/dm ³	9.0	-	2.89	-

Table 1. Average values of selected quality indicators of the concentrate from landfill leachates treatment by reverse osmosis [9, 15, 30, 32]

Table 2. Concentrations of selected eleme							
treatment of landfill leachates by reverse osmosis [mg/dm ³] [30]							

Indicator	Value	Indicator	Value	Indicator	Value
General phosphorus	23	Nitrate nitrogen V	5,620	Nitrate nitrogen III	5,388
Calcium	532	Aluminium	1,450	Arsenic	0.52
Magnesium	498	Zinc	0.8	Nickel	0.37
Potassium	3,783	Manganese	1.17	Chrome	0.2
Sodium	3,914	Copper	0.14	Lead	0.13

3. TREATMENT OF THE CONCENTRATE FROM THE MEMBRANE SEPARATION PROCESS OF LANDFILL LEACHATES

Due to the fact that membrane filtration processes do not degrade pollutants but only separate them, the separated concentrate contains most of the components separated from wastewater and should be neutralized or purified. The following methods of concentrate treatment are listed in the literature [6, 32, 37]:

- recirculation,
- incineration and solidification with other materials,
- evaporation,
- granulation,
- purification to remove impurities,
- utilization of the landfill in the block.

The technology of solidifying concentrates with materials such as fly ash or sludge from sewage treatment plants and recirculation to the landfill can be used to improve the efficiency of biochemical degradation and accelerate the immobilization of mobile components. However, due to low costs, the latter option is currently the most widely used. Proponents of this solution indicate that the landfill can be compared to a bioreactor in which biogas is produced. This requires appropriate conditions with respect to humidity and air in the landfill to accelerate biochemical processes and shorten the time of biochemical transformation as well as migration of nutrients and microorganisms in the landfill. In addition to recycling to the landfill, the concentrate can be subjected to evaporation and the resulting residue directed for storage. The concentrate granulation technology is also used [37]. It is an energy-consuming technology consisting of drying concentrate in hydrocyclone devices with steam heating and a distillate cooler. Concentrated leachates are directed to a special dryer, where dry granules are formed. Post-process sewage, created after drying, is discharged into the leachate reservoir, while the resulting granulate - deposited in a landfill. The disadvantage of the process is the reintroduction of impurities to the waste mass. In the described cases, the problem is the increase in the concentration of toxic compounds in the body of the landfill, which are not biodegradable and accumulate in the system. The literature presents works on the impact of concentrate recirculation on the quality of leachates and indicates an increase in the value of COD, ammonium nitrogen and an increase in salinity in raw landfill leachates. As a consequence, this negatively affects the efficiency of the RO installation itself and reduces the value of impurity retention coefficients [6, 24]. Another solution to the concentrate problem is their disposal or economic use. For the disposal or further treatment of concentrates from the process of membrane separation of leachates, advanced chemical oxidation, electrocoagulation or anodic oxidation processes can be used . The effectiveness of these methods depends on the properties of the compounds removed and the conditions of the process.

3.1 ADVANCED OXIDATION PROCESSES

Advanced chemical oxidation (AOP) processes include chemical, photochemical, catalytic and electrochemical oxidation, and combinations thereof. The most commonly used processes include the use of ozone, hydrogen peroxide or Fenton's reagent supported by UV radiation. These processes can be supported by ultrasound and/or hydrodynamic cavitation and electric current. Radicals, mainly hydroxyl OH• play the most important role in these processes. They are characterized by high oxidation potential and therefore difficult to decompose organic compounds can be completely mineralized or transformed into biodegradable compounds. Also complex hydrophobic aromatic and/or

aliphatic compounds are converted to hydrophilic compounds. Virapan et al. [40] studied the distribution of organic pollutants from the retentate resulting from the treatment of pharmaceutical wastes (pharmaceuthics Phs) and personal care products (personal care products PCP). During chemical oxidation, the efficiency of removing organic impurities from the retentate ranged from 40 to 50%. Chemical oxidation support through UV exposure resulted in a significant increase in the degree of removal of impurities from 80 to 100% [40]. Koc-Jurczyk and Jurczyk used Fenton's reagent to neutralize concentrate purification after reverse osmosis from leachates [15]. The effect of pH on the removal efficiency of organic and humic compounds was analyzed. As other authors also indicate [4, 8, 15], the best purification effects are obtained during oxidation in an acidic environment (pH 3). The efficiency of organic compounds decomposition can reach 80, 90% for COD and BOD₅ respectively At the same time, there is an increase in the value of the ratio of these indicators confirming the increase in biodegradability of the obtained product. The authors confirmed that the only operational problem with the use of Fenton's reagent is the need to acidify the reaction environment [15]. Comparative studies of the classical Fenton process and the electro-Fenton process were conducted by Fernandes and others [9]. As in the studies described above, it has been shown that higher efficacy is achieved after acidification of the retentate (pH 3). It has also been shown that the efficiency of removing organic compounds is directly proportional to the increase in current flowing through the electrodes. Process support had an impact on the increase in the amount of hydroxyl radicals, which determine the oxidation of organic compounds. However, the decrease in COD was not greater than 50%. Comparative studies of selected AOP methods, including ozone, UV radiation, hydrogen peroxide, electrocoagulation and electrochemical oxidation were conducted by Gautam et al. [10]. The research was carried out with the use of a synthetic concentrate that may be formed during the treatment of leachate from a hazardous waste landfill. In this concentrate in high concentrations there were refractive organic (chloroorganic) compounds, ammonium nitrogen, selected heavy metals and inorganic salts. Studies have shown that when using the above processes, the COD value decrease did not exceed 60%. Similar comparative studies of the effects of UV radiation, the electro-Fenton process and photo-electro-Fenton process on the removal of impurities were conducted by Asaitharnbi et al. [2]. The dependence of the removal of organic compounds expressed by the COD indicator on the initial concentration, pH value, colour, hydrogen peroxide concentration, electricity consumption and UV dose was determined. The test results showed that under the most favourable process conditions of the hybrid photo-electro-Fenton process, removal of organic compounds at the level of 97% and complete colour removal was achieved [2]. Photochemical catalytic oxidation of organic compounds in reverse osmosis concentrate has been studied by Westerhoff et al. [41]. The combination of UV radiation in the presence of a catalyst, which was titanium dioxide (UV/TiO₂), allowed to obtain a high degree of removal of organic compounds. The inclusion of filtration in AOP has contributed to a higher efficiency in removing organic compounds at lower UV doses. At the highest dose of UV radiation (10 kWh/m³), the concentration of dissolved organic carbon in the RO retentate decreased by 80%, while in the case of the integrated process (with filtration) it reached 91% [41].

3.2 ELECTROCOAGULATION AND ANODE OXIDATION

Electrocoagulation (EC) [5, 7, 13, 18, 23, 34] is an effective method of purifying leachates from landfills, allowing the removal of dissolved and undissolved impurities without introducing additional reagents. This process can also be used at a later stage of purification of both permeate and concentrate. Studies by Gautam and others have shown that the use of EC allows the removal of organic compounds determined in 50% while reducing the content of heavy metal ions to 90% [10].

Malinovic et al. conducted research on concentrate disposal from the RO process in the electrocoagulation process [21, 22]. The experiment was carried out using anodes made of iron or aluminium, in a batch electrochemical reactor. Changes in the concentration of dissolved compounds (determined as a dry residue) and turbidity were monitored. The research results showed that with the use of the iron anode, the efficiency of removing dissolved compounds and turbidity was 96 and 22%, respectively, while with the aluminium anode - 17 and 11% [21, 22].

Fernandes et al. Also investigated the possibility of using EC to neutralize retentate А comparative reverse osmosis [9]. experiment using electrocoagulation and anode oxidation (AO) and the Fenton process were carried out in parallel. Tests were carried out in periodically powered reactors. During electrocoagulation, stainless steel electrodes were used at variable currents. The removal of organic compounds expressed as the COD index depended on the current and ranged from 32 to 65%. Anode oxidation was carried out under the same conditions, but using a two-cathode system between which the anode was mounted. The cathodes were stainless steel plates and the anode was a bipolar electrode. The efficiency of removing organic compounds designated as COD in the AO process depended on the pH value of the reaction medium and the intensity of the current. With the decrease of the pH value and the increase of the current intensity, the oxidation efficiency increased and reached the maximum value of 74% [9].

A relatively new solution is the patented technology with the use of electrode concentrator (EDC) [3, 11]. In this reactor, a graphite anode was designed, while

a cathode was made of stainless steel. In addition, cationic and anionic membranes were placed alternately in the reactor at a distance of 1.5 mm, obtaining desalination and concentration chambers. Introduction of sodium chloride solution into the electrode chamber and application of voltage causes ion migration through ion exchange membranes and concentrate concentration. The research was carried out using retentate after reverse osmosis, which is formed during leachates treatment. The removal efficiency of organic compounds expressed as COD index reached 70%. Moreover, ammonium nitrogen concentration was reduced by 45%, total iron - 97%, total hardness - by 42% and colours by 74%. The disadvantage of this technology is the increase in chloride concentration by about 66% and the formation of deposits. However, the brine obtained after the process can be electrochemically transformed into sodium hypochlorite and reused for decontaminating landfill leachates [3, 11].

4. USE OF CONCENTRATE

As already mentioned, qualitative studies of the concentrate from membrane separation are few. Based on the available data, it can be concluded that the qualitative composition is diverse, which makes it difficult to develop the conditions for use. The high concentration of organic compounds, nitrogen compounds and the presence of potassium in leachates mean that they can be used to intensify the cultivation of industrial plants. However, it should be taken into account that on the one hand the concentrate contains fertilizer components, and on the other - toxic compounds that can accumulate in soil and organisms. Thus, such management of concentrates creates the danger of leaching of toxic components and migration to soils and the aquatic environment. Initial research in this area was conducted by the Co-author [29]. The experiment used diluted landfill leachates, which was used to intensify the cultivation of energy willow (Salix viminalis). In the conducted experiment, in determined conditions, an increase in energy willow growth was noted to 70%. From a logistical point of view, when industrial cultivation is significantly distant from the municipal waste landfill, it seems more convenient to use a concentrated concentrate for this purpose after RO. Another solution would be to establish a willow plantation near the landfill, which could further enhance the landscape.

5. CONCLUSION

Reverse osmosis modules are often used in landfill leachates treatment installations, from which a concentrate of various qualities is discharged. The volume of this concentrate accounts for 25% of the stream directed for purification, so this is a significant amount. An additional source of sewage is

the chemical cleaning of membranes, which is necessary due to the varying qualitatively leachates and high membrane separation capabilities, which promotes phenomena such as fouling or scaling. The problem with concentrate and sewage generated after membrane cleaning processes is not entirely solved. Usually these streams return to the waste mass. Due to the disadvantages of concentrate recirculation to the body of the landfill, consisting in enrichment of waste with concentrated components previously separated from leachates, it is necessary to search for other solutions. Among the processes of neutralizing retentate, in-depth oxidation and electrocoagulation are the most effective. AOP methods allow for effective degradation of organic compounds and increase of biodegradability of residues. However, it requires the use of chemical reagents and possibly UV power supply to the system. The electrocoagulation process is also associated with energy demand. The comparison of processes in economic terms is not easy because it depends on the concentration and type of pollution as well as the type and technological parameters of the disposal process. In addition, energy consumption, and hence the cost of treatment, increases as the quality requirements of the disposal product increase. Economic use seems to be another solution. An example would be growing energy crops. However, such application requires additional research not only in terms of assessing the impact on the growth of plant mass but also in terms of assessing the impact on soil and water environment".

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