Treatment of municipal waste landfill leachate with low pressure reverse osmosis and nanofiltration membranes

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Abstract. Two different approaches to treat municipal landfill leachate with reverse osmosis membranes are evaluated and discussed. Possibilities are evaluated to radically reduce concentrate flow by a value that does not exceed 0.5-1 % of initial feed water flow to withdraw all rejected impurities together with dewatered sludge. Experimental procedure is developed and described to evaluate reduction of membrane flux and rejection during leachate treatment and recovery increase. Results of experimental investigations are presented that enable us to determine main characteristics of membrane process such as: membrane flux, ammonia rejection, membrane types, number of stages, and recovery. Experimental relationships are obtained to determine the required membrane recovery values that correspond to ammonia concentration in the feed water to meet required regulation values in the product water.

1. Introduction
Construction of landfills is a conventional solution for municipal solid wastes disposal and storage [1]. Generation of leachate by municipal solid waste landfills causes a significant threat to surface and ground water. The landfill leachate contains many organic and inorganic contaminants generated within landfill body [1]. Different chemical and biological methods are applied to remove these contaminants [2]. To remove dissolved salts and biogenic elements (like ammonia and phosphate ions) reverse osmosis (RO) membranes are applied [3]. Often leachate that has high TOC value (from 1000 to 5000 ppm) is subjected to chemical treatment to remove suspended matter and dissolved organics [4]. High organic content increases osmotic pressure and decreases membrane permeability [5]. From the other point, chemical treatment, coagulation, flocculation, and further sedimentation require large volumes of water treatment structures and footprint. The paper is aimed at providing experimental results to develop efficient membrane techniques and evaluating required membrane area and operational costs. Figure 1 shows a flow diagram of landfill leachate handling and treatment.

RO is widely applied in many wastewater treatment projects [4, 5] due to membrane abilities to reject inorganic (ammonia, phosphates, and chlorides) and organic (humid acid, halogenocarbons, and phenols) contaminants [5, 6]. Often a double-stage membrane treatment technique is required to reach low concentration values of nitrates and ammonia in the product water (Fig. 2) to meet strict water discharge regulations [2, 5]. When landfill leachate is purified using membrane techniques, the problem of concentrate handling and utilization often arises [6]. Concentrate flow usually constitutes 15-30 % of leachate volume [5, 7]. In landfill leachate treatment practice, concentrate is returned back to landfill body [2] that can cause increase in different contaminants concentration values with time.
Figure 1. Balance diagram of landfill leachate handling (100 cu. metre per day capacity).

Figure 2. A balance flow diagram of landfill leachate treatment using a two-stage RO process with concentrate utilization: 1 – feed tank; 2 – first stage pump; 3 – RO membranes; 4 – nanofiltration membranes (third stage) to reduce concentrate flow; 5 – second stage product tank; 6 – second stage pump; 7 – second stage RO membranes.

In [8], authors have presented a new technique to reduce concentrate flow to reach 0.3-1.0 % of initial feed water flow that enters membrane facility. Thus, concentrate can be withdrawn and utilized together with dewatered sludge [8, 9]. In present paper, authors aimed at the application of approach described in [8] to treat landfill leachate and to produce quality desalinated water and dewatered sludge that can be utilized using verification, incineration, or other techniques. The flow diagram of the process of radical concentrate flow reduction and withdrawal together with dewatered sludge is shown in Figure 3.

To pretreat leachate prior to RO desalination, chemical treatment techniques were applied [3]. As it was discussed in [5, 8], the presence of high molecular organics in the feed water decreases membrane flux. Sedimentation of coagulated and flocculated organics provides additional amount of sludge that
facilitates concentrate utilization: the sludge moisture value of 80 % corresponds to water amount in the sludge five times more than amount of solids. Therefore, high TOC values in leachate provide favourable conditions to use the developed techniques [5]. For the cases where leachate is directly treated by membranes, research was aimed maximum recovery values to return minimum water back to landfill body (Fig. 1).

Present study demonstrates results of experimental evaluation of characteristics and parameters of RO process to purify leachate. As concentration values of ammonia in leachate vary from 80 to 2000 ppm, two stages of RO membrane treatment were applied (Fig. 2).

2. Materials and methods
The test unit flow diagram is shown in Figure 4. Wastewater after bioreactor was collected in the feed water tank 1. The pump 2 to the module 3 pumped wastewater. Concentrate was returned back to the tank 1 and product flow was forwarded to product tank 4. Pressure value was regulated using the pressure valve 5 and controlled by pressure gauge 6. RO membrane used were 1812 type elements made by CSM Company, Korea (model 1812 BLN). For concentrate treatment, 90NE membranes were used (model 1812 90NE). The gear pump created pressure. Pressure value was 6 Bars.
Chemical compositions of landfill leachate without chemical treatment, after chemical treatment, after the first and the second stages of RO are presented in Table 1.

Table 1. Results of chemical analyses of leachate after treatment with chemicals and RO membranes.

| Parameter          | Leachate (feed water) | After flocculation | After RO first stage | After RO second stage |
|--------------------|------------------------|--------------------|----------------------|-----------------------|
| COD, ppm           | 1728                   | 605                | 15                   | 8                     |
| pH                 | 8.9                    | 6.5                | 6.8                  | 6.2                   |
| Hardness, meq/l    | 15.5                   | 8.8                | 0.3                  | 0.05                  |
| (milliequivalents   |                        |                    |                      |                       |
| per litre)         |                        |                    |                      |                       |
| Alkalinity, ppm    | 76.0                   | 13.6               | 5.0                  | 2.1                   |
| Ammonia, ppm       | 425                    | 92                 | 3.1                  | 0.5                   |
| Nitrate, ppm       | 65.5                   | 42.1               | 5.85                 | 2.73                  |
| Chloride, ppm      | 2308                   | 265                | 27                   | 2                     |
| Sulphate, ppm      | 627                    | 508                | 0.68                 | 0.12                  |

3. Results
Experimental data were consistently processed to obtain values of the main technical parameters of membrane unit. First of all, dependences of various ions concentration values on the recovery and the product volume were determined. Figure 5 shows dependences of ionic concentrations of sulphates, chlorides, and ammonia ions on concentration factor K values, where K is a ratio of the initial volume in tank (1) (Fig. 4) to the volume of product water in product tank (4) during the test run.

At the beginning of experiment, initial volume was 70 litres. This volume was treated on the first stage of experiment by low pressure RO BLN type membranes until the amount of water in feed tank (1) (Fig. 4) was 7 litres and the volume of product water collected in tank (4) was 63 litres. On the second stage of experiment, the remained amount of concentrate (7 litres) was treated using nanofiltration 90-NE membranes that demonstrated higher product flow.

![Figure 5.](image)

**Figure 5.** Concentration values of different species and COD concentration values in RO concentrate (a) and RO product water (b) versus K: 1 – chlorides; 2 – ammonia; 3 – COD after chemical treatment; 4 – sulphates; 5 – COD w/o chemical treatment; 6 – chlorides, second stage; 7 – ammonia, second stage; 8 – sulphates, second stage.
Then, decrease of membrane product flow throughout the experiment was evaluated as a dependence on K value (Figs. 6-10).

Figure 5a shows dependences of ammonia and sulphate ionic concentrations as well as COD (chemical oxygen demand) values in concentrate flow during experimental test run. At different stages (that corresponded to certain K values, such as: 2, 4, 6, 10, 20, 40, 80), product water samples were withdrawn where concentrations of all impurities as well as TDS values were determined.

Similar dependences of different species’ concentration values in product water and concentrate versus K are shown in Figure 5. Figure 5a shows increase in different rejected by membrane impurities versus K.

![Figure 6. Results of evaluation of concentrations of different species in RO and NF product water versus K: 1 – chlorides; 2 – ammonia; 3 – COD after chemical treatment; 4 – sulphates.](image)

![Figure 7. Reduction of specific membrane flux as a function of coefficient K value: 1 – product flow without chemical treatment, BLN and 70NE membranes; 2 – product flow after chemical treatment, BLN and 70NE membranes; 3 – product flow after chemical treatment, 70 NE nanofiltration membranes.](image)
As it is shown in Figure 5b, ammonia concentration values in product are higher than discharge regulation values. This can be explained by the low rejection of monovalent ions by low-pressure RO membranes in the pressure range of 6-12 Bars. Ammonia ion rejection in this pressure range varies between 85 and 90 %. Therefore, to arrange high rejection of ammonia, the second stage RO BLN membrane treatment should be applied.

Figure 6 shows increase in different species concentration values on both stages of concentrate treatment. Reduction of specific product flow as a function of coefficient K value is shown in Figure 7. Product flow was measured under pressure value of 6 Bars.

4. Discussion
The next step was devoted to determination of technical parameters of membrane unit. Experimental data processing was aimed at determination of technical characteristics of membrane unit, such as membrane area required and minimal concentrate amount. To predict influence of membrane recovery on product water quality, various relationships were examined.

Figure 8. Determination of specific membrane flux values within various ranges of K and product volumes, influence of TOC on flux and membrane area: product flux versus K (a) and product flux versus product volume (b).
Figure 9. Determination of RO and nanofiltration membrane area required to reach K values at the first and second stages of membrane unit to treat 1000 l of leachate per hour: a) specific membrane area required to produce calculated volumes of product within different K intervals at the first stage; b) BLN membrane area required to produce calculated K values within different K intervals at the first stage; c) specific membrane area to produce calculated volumes of product within different K intervals at the second stage; d) 90NE membrane area required to produce calculated product volumes within different K intervals at the second stage.
Main steps to calculate the required membrane area and amount of membrane modules during the first stage of water treatment with BLN membrane when K is increased from 1 to 7 is illustrated in Figure 8. Figure 8a shows the amounts of product obtained in the K ranges from 1 to 3, from 3 to 4, and from 4 to 7. The obtained product amounts are presented as a percentage of the total volume produced during treatment of 70 litres of the feed wastewater with the final K value of 7 (after 60 litres of product water was produced). The selection of value 7 is explained by the low product water quality obtained with K values from 7 to 10. Figure 8b presents results of determination of membrane product flow rates in different K ranges. The calculations are made for the case when the required product flow of membrane facility is 1000 litres per hour and concentrate flow is 140 litres per hour. For each range of change of K value product flux was determined and the arithmetic average value of membrane specific flux was determined, expressed in: litres/hour / square meter (Fig. 8).

Using calculated values of product water produced during one hour in the selected range of K and average specific value of membrane flux within this range, we can calculate the required membrane area to provide the required amount of product water during one hour (Fig. 8). Specific product flow decrease in BLN and 90 NE membranes versus K is shown in Figure 9a. Figure 9b shows results of membrane area determination within the selected ranges of K variation. Calculations are made for the case when BLN membranes treat wastewater and K value changes from 1 to 7, and the pressure was 6 Bars. The required number of membrane elements can be determined assuming that membrane area in a standard 4040 element (BLN 4040) manufactured by CSM Company (Korea) equals 10 square meters. Thus, to decrease wastewater flow by 7 times and to produce 1000 litres of quality product water per hour, we need to use 12 elements of 4040 standard. The K value grows "step by step"; therefore, elements can be connected in series: 5 in parallel, then 4 and 3.

![Figure 10](image.png)

**Figure 10.** Evaluation of the required membrane area to treat 1000 litres of leachate per hour and to reach the required recovery (or K) value, the calculated membrane area required to reach different K values within different K intervals: 1 – leachate after chemical treatment (BLN and 70 NE membranes); 2 – leachate without chemical treatment (BLN and 70NE membranes); 3 – treatment of leachate after chemical treatment with nanofiltration 70NE membranes.

Figure 10 demonstrates similarly obtained results of calculation of required membrane area to evaluate a number of membrane elements calculations required to further reduce concentrate flow from 140 to 10 litres per hour on the second stage of concentrate treatment. Reduction of membrane product flow on the first and on the second stages of treatment (both for RO and nanofiltration membranes) versus K values is shown in Figure 10. For the second stage, 90NE nanofiltration membranes were used. As it is shown in Figure 10, to reduce concentrate flow on the second stage, 50
square meters of membrane surface is required (5 membrane elements 90NE 4040 are needed connected in series: 3 in parallel and 2 in parallel).

Both cases of treatment of leachate without chemical treatment and after it are determined. For the case when leachate was not subjected to chemical treatment and is directly purified by RO membranes, the first stage recovery value did not exceed 90% due to the loss of product flow. Figure 10 demonstrates results of membrane area calculations as a dependence of specific required membrane area (to treat 1000 litres per hour of leachate) on K value reached during leachate treatment.

A 4040-type membrane modules connection diagram is presented in Figure 11. For the case, when leachate is treated by chemicals and organics is coagulated and deposited prior to RO treatment, RO membranes on the first stage provides decrease in feed water flow by 6-10 times (Fig. 11a). Concentrate after the first stage is collected in the intermediate tank where additional chemical treatment is applied to further deposit organics reduce COD value.

![Figure 11. Results of the required membrane area evaluation for the first and the second stages of membrane unit (1000 l / h capacity) and membrane module connection flow diagram: 1 – first stage pump; 2 – membrane module BKN 4040; 3 – membrane module BKN 4040; 4 – second stage pump; 5 – membrane module 70 NE 4040.](image-url)
After chemical treatment, concentrate is treated on the second stage using 90 NE nanofiltration membranes to reach high recovery values up to 98-99%. For the case when chemical pretreatment of leachate is not implemented, high recovery values are not reached due to high COD of leachate and low membrane flux (Fig. 11b). More 4040 BLN membrane elements are used to produce purified water and concentrate after BLN modules directly enters 90NE nanofiltration modules to further reduce concentrate flow to reach 90% recovery value.

Figure 12 shows a flow diagram of landfill leachate treatment. To remove suspended matter prior to RO membrane treatment, sedimentation of suspended matter in tank 3 and further filtration of clarified water using filters 6 is applied. TOC value does not change during pretreatment. This approach is used when leachate is “directly” treated by RO. To radically reduce TOC, coagulation and flocculation are applied using high stoichiometric concentrations of chemicals (ferric chloride, etc.). After pretreatment, leachate enters the first stage of RO treatment with recovery value up to 85-90%.

Figure 12. Landfill leachate treatment flow diagram: 1 – wastewater pump; 2 – leachate collection tank; 3 – sedimentation tank; 4 – clarified water tank; 5 – pump to filters; 6 – filters; 7 – filtered water pumps; 8 – RO system pump; 9 – RO membrane; 10 – nanofiltration membrane for recovery increase; 11 – concentrate collect tank for sedimentation (optional); 12 – clarified concentrate tank; 13 – third stage pump; 14 – third stage membrane; 15 – sludge «thickening» tank; 16 – clarified water recycling pump; 17 – sludge pump to dewatering; 18 – «bag» dewatering unit; 19 – dewatering reject pump; 20 – second stage RO pump; 21 – second stage membranes; 22 – first stage product pressure tank; 23 – second stage tank.

To facilitate further increase in recovery during the third stage of nanofiltration increase, leachate first stage concentrate is collected in tank 11 where coagulation and flocculation of organics occurs. Coagulation and sedimentation of deposited organics required large volume of water treatment structures and footprint. For the case when leachate is "directly" treated by RO, the intermediate tank 11 and the third stage pump are not used.

5. Conclusion
Analysis and studying of the experimentally obtained results allowed us to draw to the following conclusions.

To reach the required ammonia concentration in product water, double stage treatment of feed with low pressure reverse osmosis membrane treatment is required. High recovery values are reached through the additional treatment of concentrate with nanofiltration membranes.
Influence of dissolved organics defined as COD on membrane performance is investigated. Leachate COD decreases membrane flux and should be accounted when membrane area is determined. Flocculation and further sedimentation of organics prior to reverse osmosis treatment facilitates membrane filtration and provides higher values of recovery.

Application of reverse osmosis and nanofiltration membranes combined with chemical leachate pretreatment enables us to produce quality water that meets discharge regulation standards and concentrate flow that does not exceed 0.3-0.5 % of the total feed water that enters water treatment plant.

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