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Reduction of Ecological Damage From Landfill Leachate Using Reverse Osmosis Technology with Concentrate Utilization

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Abstract: Landfill leachate is currently recognized as a source of serious environmental danger. Reverse osmosis is an effective method for treating landfill leachate to comply with wastewater discharge standards. However, this technology generates a significant volume of concentrated waste, which is expensive to manage. This research explores methods for producing high-quality water suitable for industrial applications or discharge into surface water sources while minimizing the concentrate output. The findings demonstrate the potential to drastically reduce concentrate flow to a mere 3-4% of the initial feed water volume. According to developed technology, concentrate is hidden in the sludge transported to the landfill. Is achieved due to application of the developed «ion separation» technology. The presented technology allows to reduce concentrate discharge to economically reasonable values to apply evaporation or withdrawal of concentrate by motor transport. It is possible to achieve a reduction in concentrate consumption by changing the ratio of concentrations of salt ions and organic substances. Such separation of concentrate solution can be implemented due to the duse of membranes with different rejection of divalent ions and organics. The achieved separation of the concentrate solution enables us to significantly reduce the costs of its utilization.

1 INTRODUCTION

1.1 Landfill leachate: ecological hazar

The use of reverse osmosis membrane technology to purify landfill leachate allows to prevent contamination of surface and ground waters with decomposition products of organic substances. Application of reverse osmosis technology provides an economically acceptable solution of the problem compared to the use of evaporation or biological methods, as well as chemical precipitation techniques.

Meanwhile, reverse osmosis technology has a number of disadvantages that prevent it is wide industrial application. The first problem is high operation costs associated with high energy and reagent consumption. And the second problem is the concentrate handling the flow rate of which is 25-30 percent of the total amount of leachate treated by membrane plant.

Drying concentrated wastewater by evaporating water to obtain solid salts is considered a highly effective method for managing concentrated waste streams. However, the presence of organic compounds hinders both water evaporation and salt crystallization. A more significant reduction in concentrate volume can be achieved through separating organic and inorganic components. This is due to the fact that solutions with organic compounds have an osmotic pressure that is 4 to 5 times lower than that of electrolyte solutions at the same concentration. By separating the concentrate into distinct organic and salt solutions, we can significantly reduce the overall volume and lower

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disposal costs, as each component can be treated independently and more effectively.

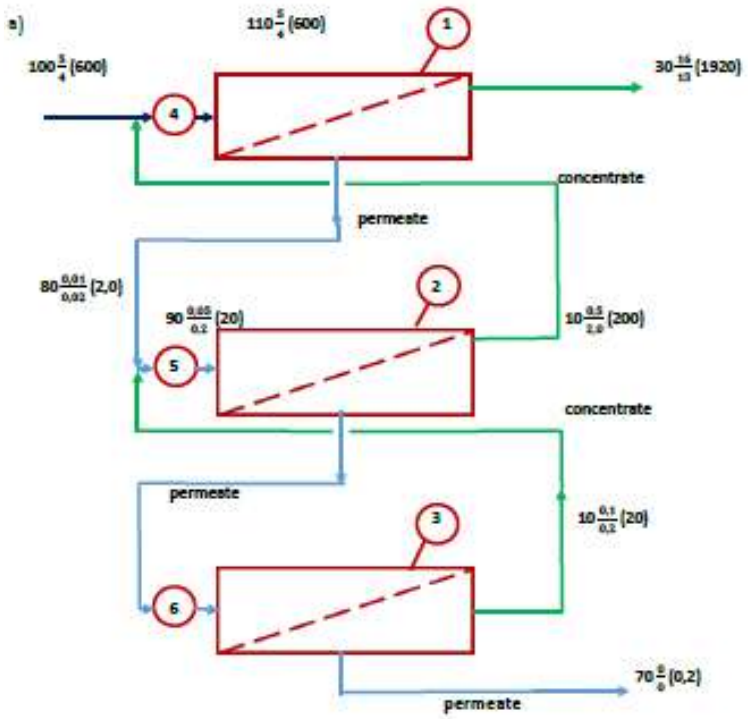
Adjusting the proportions of salt and organic material within a solution can lead to a decrease in concentrate usage. The new technology developed by the authors utilizes separation of concentrate solution into two concentrated solutions of salts and organic substances. Such separation allows to treat separately and reduce the volume of each solution at lower costs.

Currently, a number of landfills are allowed to discharge concentrate back into the body of the landfill, but this temporary measure leads to increased concentrations of contaminants in the leachate and further increases the cost of treatment. Evaporation plants are also used for concentrate utilization at a number of landfills, but a way to reduce the volume of concentrate sent to evaporation must be found to successfully and economically solve the problem.

The present work describes a new approach to the development of membrane plants with reduced concentrate consumption - no more than 3-4% of the total volume of landfill leachate.

1.2 Existing technologies to reduce and utilize concentrate

Fig. 1(a) describes a conventional approach to treat landfill leachate using high pressure and high rejection reverse osmosis membranes usually applied at sea water desalination projects. This scheme allows to provide high quality product water but fails to obtain a concentrate flow rate less than 30% of the initial leachate fed to the reverse osmosis facility.



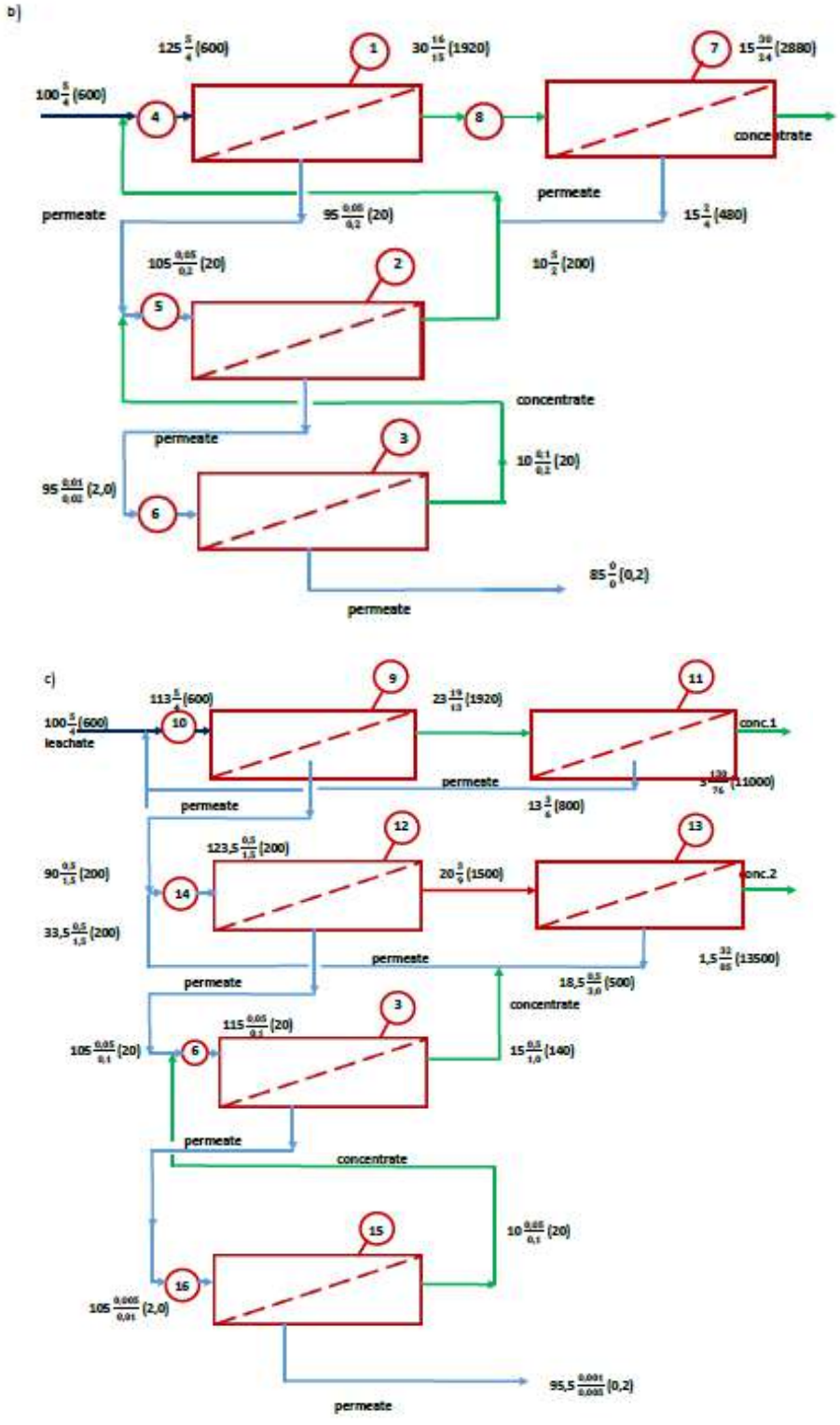


Figure 1: Flow diagrams of reverse osmosis membrane facilities used to purify landfill leachate: a) technology that uses high pressure «seawater» reverse osmosis membranes; b) a conventional approach to reduce concentrate discharge: the use of high pressure additional membrane stage; c) a new approach to reduce concentrate: the use of low pressure low rejection nanofiltration membranes to increase recovery.

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The scheme shown on then Fig. 1(b) is a «modernized» scheme of conventional scheme (Fig. 1 (a)), that includes additional high-pressure membrane modules to reduce concentrate consumption. Globally, "filter-press" membrane modules equipped with high-pressure reverse osmosis membranes produced by Rochem (Germany) have proven to be the most effective and dependable solution for treating landfill leachate. This technology allows to deionize solutions with total salt content of 80-90 grams per liter using working pressure value up to 15 MPa. This approach is used at a number of landfills in the Moscow region for modernization of leachate treatment facilities to reduce concentrate discharge by more than two times.

In landfill leachate treatment practice a lot of measures were applied to reduce concentrate flow and increase concentration ratio. The possibility to increase concentration of salts was reported in that consisted of the use of " cascade" of nanofiltration membranes with low rejection. This allows to achieve concentration values up to 150-180 grams per liter using low working pressure 16-18 Bars (1,6 - 1,8 MPa).

The scheme presented on the Fig 1(c) presents the new technology developed by the authors, which allows to achieve high concentration ratio of the initial leachate by applying a "cascade" of nanofiltration membrane modules. However, the high content of organic matters in the concentrate streams on the first and on the second stages of membrane facility makes the use of concentrate evaporation ineffective. Concentrate contains organic matters that prevents salt crystallization during evaporation as it inhibits crystal formation and prevents dry salt production.

1.3 Principles to further reduce concentrate discharges

Reverse osmosis plants treating landfill leachate with high dissolved salt content often face a challenge: the presence of dissolved organic compounds in the concentrated brine byproduct. These organics elevate osmotic pressure, diminishing the membrane's permeability and hindering effective volume reduction. Moreover, they interfere with salt crystallization during evaporation, preventing the production of dry salts. Consequently, the presence of organics significantly impacts operational costs [11-12]. Separating organics from salts offers a solution. It allows for substantial concentrate reduction without increased energy consumption because organic solutions exert 4-5 times lower osmotic pressure than electrolyte solutions at the same concentration [14]. Below principles to reduce concentrate floe using its separation in organic solution and salt solution are disclosed. For an example the landfill leachate chemical composition is taken from the Table 1 where COD of the landfill is 5000 ppm and TDS is 4000 ppm. For the case when we deal with salt solution with TDS value of 4000 ppm without organics, we can achieve 60000 ppm salinity in concentrate by applying high pressure reverse osmosis membranes and using working pressure of 60 Bars (Figure 1,a). We can also apply "cascade" of low rejection and low pressure nanofiltration membranes and reach concentration of salts 160-180 gram per liter at a working pressure value of 5 MPa (Figure 1,c). Thus, in the first case initial feed water salinity is increased by 15 times and in the second case it is increased by 40-45 times. Principles of concentration value increase using reverse osmosis method is based on the assumption that the required pressure value is evaluated not by the concentration value of separated solution but by the difference between osmotic pressure in concentrate and permeate. The higher is salinity of the feed water, the lower is the rejection value of membranes. The feed water solution (landfill leachate) with 4000 ppm salinity has osmotic pressure of 3 Bars, and the organic solution with COD value of 5000 ppm has osmotic pressure value of 0,85 Bars. When feed water with 4000 ppm salinity is concentrated by 15 times, we reach 60000 ppm in concentrate that corresponds to osmotic pressure value of 45 Bars. We apply working pressure of 60 Bars and reach osmotic pressure value of 45 Bars and provide a "driving force" (difference between the working pressure and osmotic pressure in concentrate) 15 Bars. In case we have organics in the feed solution, after we increase initial concentration by 15 times we achieve osmotic pressure value of (3 + 0,85) x 15 = 57 Bars that eliminates the "driving force". This explains why the presence of organics is an obstacle to reach high recoveries. Separation of landfill leachate into two concentrated solutions: salt solution and organic solution and separate further concentrating of these solutions provides substantial reduction of concentrate discharge as organic solution is better concentrated due to lower osmotic pressure.

Table 1: Chemical compositions of landfill leachate and product water after three stages of membrane treatment.

№	Ingredients	Landfill leachate	IV Stage permeate

1	COD, ppm	7500	2.4
2	Specific electric conductivity microsiemens / cm	5.5 x 10^3	34
3	Ammonia, ppm	1100	0.2
4	Total hardness, millieqivalent / Liter	27	0.2
5	Lead, ppm	< 0.001	—
6	Copper, ppm	0.0948	—
7	Zinc, ppm	1.4	—
8	Sulphate, SO4, ppm	860	1.2
9	Alkalinity, ppm	760	—

To separate organics and salts, a low rejection nanofiltration membranes are used with different rejections of organics (COD) and monovalent ions. During treatment of the test solution, the ratio of concentration values of organics and salts (COD:TDS) in concentrate increases and reduces in permeate as membranes better reject organics. After we increase the ratio value and increase COD and TDS values by 5-10 times, the concentrate should be diluted by deionized water to obtain initial TDS value. Then the second concentrating cycle should be applied where the value of COD/TDS ratio will be further increased. The third dilution/concentrating cycle provides further increase of the ratio. Figure 2 shows the balance flow diagram of three steps of COD/TDS ratio increasing. The results of osmotic pressure values calculations in concentrates and permeates on the each membrane stage.

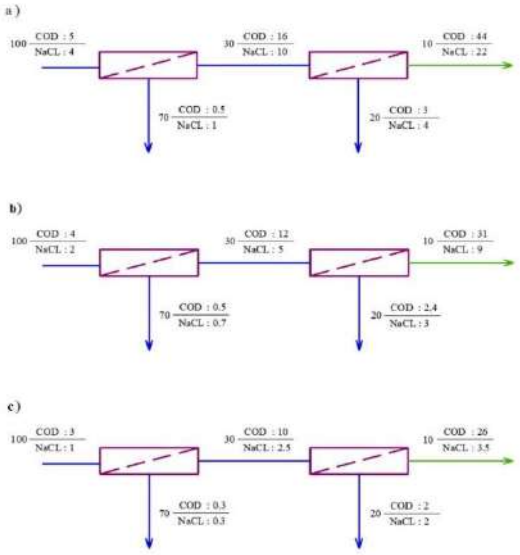

Figure 2: The flow balance diagram of the steps to separate concentrate into organic and saline solutions and to increase the COD:TDS ratio value. TDS value is 4000 ppm, COD value is 5000 ppm. a) concentrating of the feed leachate; b) dilution of concentrate by 10 times with deionized water and further concentrating; c) secondary dilution by 10 times and further concentrating.

Also, osmotic pressures of pure sodium chloride and organic solutions are presented for comparison. Basing on the results presented in the Table 2, a graph to determine the value of maximum initial volume reduction coefficient K is presented on the Figure 3. Figure 3 shows the dependencies of the «driving force» (the difference between osmotic pressures in concentrate and permeate) on the K value. These dependencies are presented for the three steps of leachate treatment (Figure 2) and for pure solutions of sodium chloride and organics (Table 2). A horizontal line parallel to the abscess axe shows the selected maximum osmotic pressure difference that should not be exceeded to provide enough «driving force» for leachate purification using low pressure low rejection nanofiltration membranes.

Table 2: Experimental results.

Ratio: Concentration, gram/liter, osmotic pressure Bar							
Indicatiors	Feed water	Permeate		Concentrate		Osmotic pressure difference, πc - πp	
		K = 3	K = 9	K = 3	K = 9	K = 3	K = 9

Solution 1 100							
NaCL, g/L	$\frac{15}{30}$	$\frac{1.7}{0.1}$	$\frac{5}{0.3}$	$\frac{38}{2.5}$	$\frac{74}{5}$	2.4	4.7
COD	$\frac{20}{0.6}$	$\frac{1.5}{0.045}$	$\frac{2}{0.06}$	$\frac{61}{1.8}$	$\frac{168}{4.5}$	1.75	4.44
π , Total Bar	1.6	0.145	0.36	4.3	9.5	4.15	9.14
Solution 2 after dilution 1:10							
NaCL, g/L	$\frac{9}{0.6}$	$\frac{1.5}{0.1}$	$\frac{7}{0.06}$	$\frac{24}{1.6}$	$\frac{45}{13.0}$	1	2.14
COD, g/L	$\frac{20}{0.6}$	$\frac{2}{0.06}$	$\frac{12}{0.36}$	$\frac{62}{1.8}$	$\frac{162}{4.5}$	1.74	6.84
π , Total Bar	1.2	0.16	0.42	3.4	7.5	2.74	8.98
Solution 3 after next dilution 1:10							
NaCL, g/L	$\frac{3}{0.2}$	$\frac{0.5}{0.03}$	$\frac{2.3}{0.02}$	$\frac{7.5}{0.5}$	$\frac{14}{1}$	0.47	0.08
COD, g/L	$\frac{20}{0.6}$	$\frac{2}{0.06}$	$\frac{12}{0.36}$	$\frac{61}{1.8}$	$\frac{168}{4.5}$	1.74	4.14
π , Total Bar	0.8	0.09	0.38	2.3	5.5	2.21	4.22
Solution after separation							
NaCL, g/L: 75	$\frac{20}{1.3}$	$\frac{3.1}{0.2}$	$\frac{15}{0.12}$	$\frac{30}{2.0}$	$\frac{70}{4.8}$	1.5	2.8
COD, g/L: 25	$\frac{80}{2.4}$	$\frac{0.4}{0.011}$	$\frac{0.5}{0.01}$	$\frac{55}{1.6}$	$\frac{165}{4.8}$	1.5	4.65
Mixture , g/L 100							
NaCL, g/L	$\frac{15}{1.0}$						

COD, g/L	$\frac{20}{0.6}$						

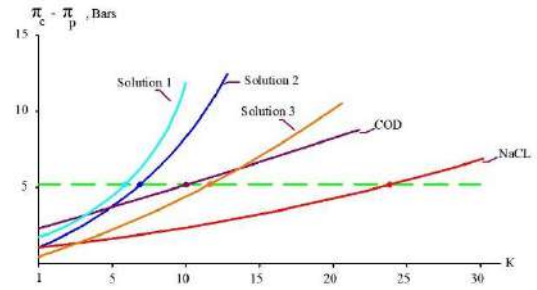

Figure 3: Evaluation of the maximum initial volume reduction coefficient K value: dependencies of the differences between osmotic pressure values in concentrate and permeate on K.

This research aimed to experimentally demonstrate the feasibility of separating landfill leachate concentrate into two distinct solutions: an organic solution and a mixture of sodium chloride and ammonium chloride solutions. The study also sought to establish recommendations for optimizing this separation process and maximizing the ratio of Chemical Oxygen Demand (COD) to Total Dissolved Solids (TDS).

2 EXPERIMENTAL PROGRAM

The experiments on separation of leachate delivered from «Voskresensk» Landfill (Moscow region) were conducted using the test membrane plant. The flow diagram of the test unit is shown on the Figure 4. The leachate feed solution was placed in the tank 1, and then directed using the pump 2 to the membrane module 3. The working pressure value was 1.3-1.6 MPa. Membrane modules of 1812 standard (1,8 inch in diameter and 12 inch in length) tailored with «nanoNF» nanofiltration membranes were used. «NanoNF» membranes were supplied by «Membranium-RM Nanotech Company» (Vladimir, Russia). The «nanoNF» membranes demonstrate low rejection of monovalent ions (50-60 per cent), and high rejection of organics (up to 90 per cent) that prompted authors to use these membranes for concentrate separation. The purified water, termed permeate, was collected in Tank 4. The remaining concentrated solution, called concentrate, was returned to Feed Water Tank 1 for further processing. The separation of organic compounds and monovalent ions took place over three stages. The first stage involved a tenfold reduction in the volume of the leachate, decreasing it from 20 liters to 2 liters.

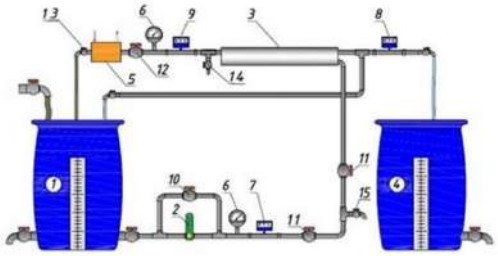

Figure 4: The flow diagram of the membrane test unit used in experimental program.

Further, on the second step, the concentrate was diluted with deionized water. Deionized water was produced by treatment of Moscow tap water by using a membrane spiral wound module (model «RE-1812 HR 75» manufactured by the company «Raifil» Russia). After concentrate was diluted by 10 times, it was again placed in tank 1 and again was concentrated by 10 times.

On the third stage the obtained concentrate was again diluted by 10 times and further concentrated by 10 times. At each stage, the ratio of COD and total salt content (TDS) in the permeate and concentrate changed continuously due to the difference in membrane rejection for organic matter and salt ions. The permeate obtained after the third stage was a mixture of ammonium chloride and sodium chloride solutions with a salt content of about 2000 mg/liter. Such solution can be effectively concentrated using reverse osmosis and nanofiltration membranes up to salt concentration of 150 g/l and higher.

This study employed rotary pumps manufactured by Fluid-o-Tec (China) operating at a pressure range of 1.2 to 1.6 MPa. Nanofiltration membranes achieved an average salt rejection rate of 60-70%, whereas reverse osmosis membranes exhibited a considerably higher rejection rate of 96%. The nanofiltration membrane modules demonstrated a specific productivity ranging from 1.5 to 15 liters per square meter per hour. The research utilized spiral wound membrane elements, including the "NME nanoNF 1812-C" model produced by Membranium

(Vladimir, Russia). Furthermore, reverse osmosis elements of the "RE 1812-HR 75" model and nanofiltration elements of the "NE 1812-70" model manufactured by Raifil (Russia) were also incorporated into the investigation. The filtration area of membrane 1812 type elements was 0,5 m2.

The research employed a variety of analytical methods to ascertain the levels of different water quality indicators. Volumetric titration techniques were used to determine the concentrations COD, alkalinity, hardness, calcium ions, and chloride ions. Photometry was utilized to measure ammonium ion and heavy metal ion concentrations, while turbidimetry was employed to determine sulfate ion concentration. Specific conductivity was assessed through conductometric analysis.

Concentrations of all species contained in leachate (total salt content, COD, alkalinity, concentrations of calcium ions, ammonia and other contaminants) of the landfill «Voskresensk» as functions of the initial volume reduction coefficient K (defined as a ratio of leachate flow rate to concentrate flow rate) are presented on the Figures 5 and 6.

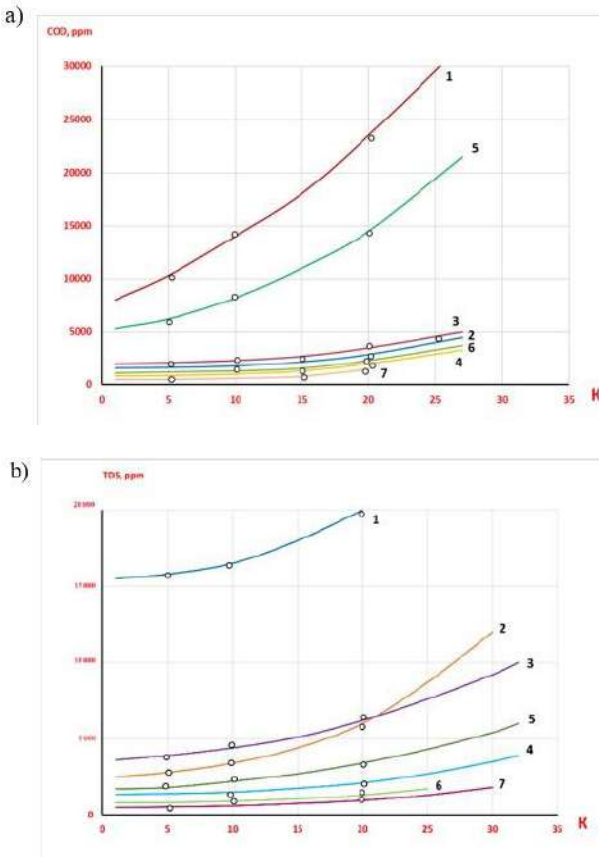

Figure 5: Dependencies of TDS and COD concentration values on K obtained in concentrates and permeates at different experimental conditions using different membranes: a) dependence of COD on K; b) dependence of TDS on K; 1 - landfill leachate treatment on the first stage using "nanoNF" membranes; 2 - first stage permeate using «nanoNF» membranes; 3 - total permeate collected on the first stage; 4 - second stage permeate using «nanoNF» membranes; 5 - the second stage concentrate using «nanoNF» membranes after dilution by 20 times; 6 - the second stage permeate using «nanoNF» membranes after dilution by 20 times; the second stage permeate produced with reverse osmosis «BLN» membranes.

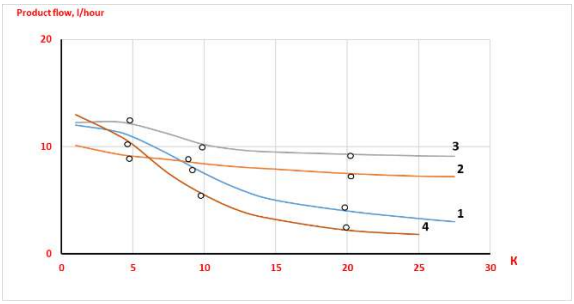

Figure 6: Reduction of membrane product flow as a function of K value at different stages of experiments: 1 - landfill leachate treatment on the first membrane stage using «nanoNF» membrane; 2 - concentrate after dilution by 10 times on the second stage using «nanoNF» membranes; 3 - the first stage permeate using «nanoNF» membranes; 4 - landfill leachate treatment on the first stage using «70 NE» membranes.

3 DISCUSSION OF THE RESULTS

During the circulation testing phase (Figure 4), the test unit demonstrated elevated Total Dissolved Solids (TDS) and Chemical Oxygen Demand (COD) concentrations, as shown in Figures 5 and 6. The buildup of dissolved organic compounds within the concentrate led to a significant increase in osmotic pressure, resulting in reduced product flow (Figure 6).The nanofiltration membranes used in the system displayed limited salt rejection capabilities, permitting salts to pass into the permeate stream. This resulted in minimal TDS difference between the concentrate and , allowing for high TDS concentrations within the concentrate (120 to 160 grams per liter) at relatively low operating pressures (not exceeding 1.6-1.8 MPa). The initial nanofiltration stage effectively removed approximately 90% of organic compounds, leading to a corresponding increase in COD levels within the first-stage concentrate. However, sodium and ammonium ions permeated through the membrane into the first-stage permeate. This permeate was then

treated in a second membrane stage using the same type of nanofiltration membranes to ensure complete removal of organic contaminants. The purified permeate from the second stage was used to dilute the first-stage concentrate before its treatment in a third stage, designed to separate organics and salts. The final stage concentrate exhibited COD levels of 120-160 g/l.

To reach high recoveries and to implement separation of organics and salts, a new flow diagram was developed by the authors (Figure 7). Untreated landfill leachate, with a daily flow rate of 100 cubic meters, a Chemical Oxygen Demand (COD) of 5000 mg/l, and a Total Dissolved Solids (TDS) concentration equivalent to 4000 mg/l NaCl, is subjected to a two-stage membrane treatment process. The first stage utilizes low-rejection nanofiltration membranes in modules 1 and 3, achieving a salt rejection rate of 50-70%. This process reduces the COD to 500 mg/l and the NaCl content to 1500 mg/l in the resulting permeate stream. The concentrate flow is significantly reduced to 3 cubic meters per day, while the permeate flow reaches 97 cubic meters per day. Subsequently, the permeates from both the first and third stages, still containing organic compounds, undergo further treatment in the second stage using low-rejection nanofiltration membranes in modules 2 and 4. The concentrate from the second stage and the concentrate from the first stage are combined in mixing tank 5. The permeate from the second stage is also directed to mixing tank 5, serving to dilute the concentrate and facilitate the separation of organic compounds.

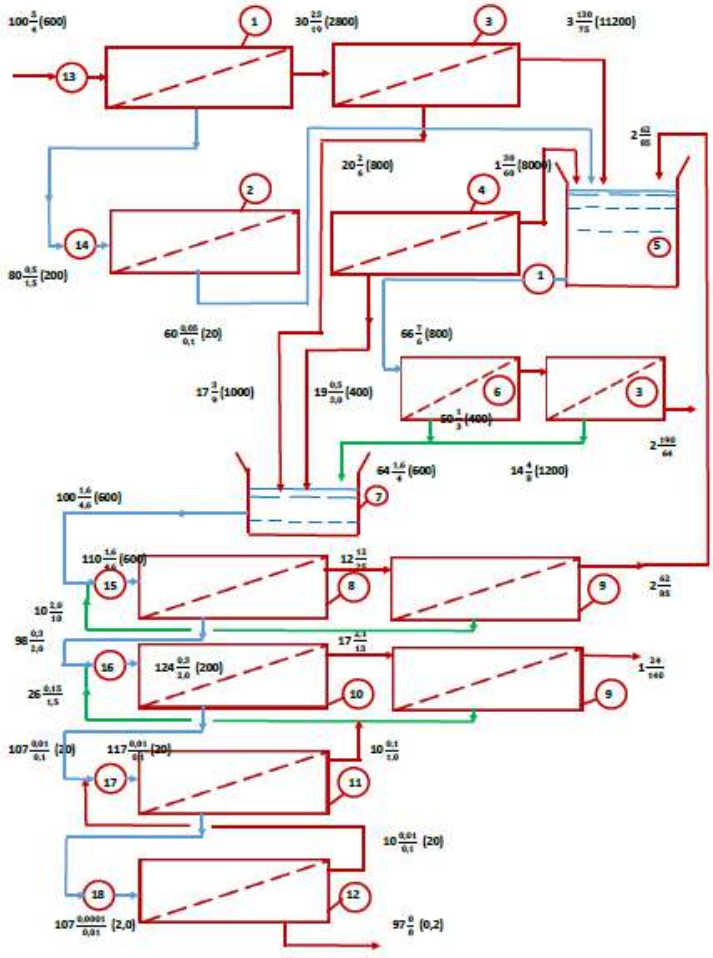

Figure 7: A flow balance diagram to separate concentrate into organic and salt solutions using the developed technology.

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The process involves diluting a concentrated solution with water in a 1:5 ratio within a mixing tank. Due to the selective nature of the membranes used, the diluted solution exhibits a higher concentration of organic compounds relative to salts compared to the original feed water. Subsequent concentration steps in modules 6 and 7 aim to maximize the COD value of the solution. This concentrated solution is then passed through nanofiltration modules 9 and 10, with the resulting concentrate being recycled back to the mixing tank. The permeate from these nanofiltration modules is further treated by reverse osmosis modules 11 to produce deionized water. Finally, a cascade of nanofiltration modules 12 is used to concentrate the residual solution from the reverse osmosis stage, achieving a target TDS value of 140 g/l.

Following an initial reverse osmosis desalination process (modules 8 and 9, Figure 7), the purified water undergoes two additional treatment stages designed to minimize ammonium concentration. Concentrates from each stage are recirculated to the preceding stage's inlet. The resulting treated water exhibits a TDS value of 5-10 ppm and an ammonium concentration of approximately 0.2 ppm. This process effectively segregates the landfill leachate into three distinct streams: final product water, saline concentrate, and organic concentrate. Firstly, the saline concentrate comprises sodium salts and ammonium at a concentration of 100-150 grams per liter, representing 2% of the initial leachate volume. Secondly, the organic concentrate contains organic compounds with a Chemical Oxygen Demand (COD) of 100 grams per liter, along with dissolved salts such as calcium, magnesium, sodium, and ammonium. This concentrate accounts for 1% of the original leachate volume. The organic concentrate demonstrates utility in landfill reclamation projects when incorporated with soil or dewatered sludge originating from wastewater treatment facilities [19-22]. The saline concentrate can be further processed through evaporation to obtain dry salts or directly utilized as a feedstock in fertilizer production.

A cost-benefit analysis was conducted to evaluate the operational expenses associated with treating leachate wastewater containing a COD of 5000 mg/l and TDS of 4000 ppm. The analysis compared conventional high-pressure membrane filtration with a novel treatment technology. Calculation of annual operating costs for landfill leachate treatment and concentrate utilization using different technological schemes is given in [23-25]. The developed technology provides lower concentrate amounts and saves a lot of money on concentrate disposal and utilization.

Figure 8: The four-stage membrane installation to treat 50 cubic meter per day of landfill leachate and to reduce concentrate by 30 times by separation into organic and salt solutions.

4 CONCLUSIONS

- This study presents findings on the quantification of both scaling and organic fouling rates. These findings are supported by scanning electron microscopy (SEM) images and spectral analyses conducted during post-mortem examinations of the membranes;
- The research findings indicate that the utilization of nanofiltration membranes effectively prevents the formation of calcium carbonate;
- This study presents economic analyses demonstrating the benefits of a novel technology employing low rejection membranes. This technology offers reduced concentrate flow compared to traditional high-pressure membrane systems, thereby showcasing its economic advantages.

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