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Evaluating Membrane Separation Effectiveness for Highly Concentrated Aqueous Media Applying Two-stage Reverse Osmosis Plant

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ABSTRACT

The research considers the issues of evaluating membrane separation efficiency for highly concentrated aqueous media by reverse osmosis. The pilot two-stage reverse osmosis plant was used for the research. The specific membrane capacity and concentration rate were chosen as test parameters determining the membrane separation efficiency for highly concentrated aqueous media. Highly concentrated leachate from solid waste landfill of chemical oxygen demand up to 10700 mgO₂/l and 400 mg·eqv/l alkalinity were used as highly mineralized water medium for testing, applying two-stage reverse osmosis plant. It resulted in determining optimal technological parameters of highly mineralized leachate processing: operating pressure for the 1-st stage reverse osmosis separation - 60 kgf/cm², concentration rate of the initial solution - by 2.9 times; operating pressure for the 2-nd stage reverse osmosis separation -15 kgf/cm², concentration rate of the 1st stage permeate - by 9 times. The research revealed the positive effect of preliminary acidification stage of highly concentrated aqueous media with high alkalinity for membrane separation process.

Key words: concentrate, concentration rate, highly concentrated waste water, membrane separation, membrane specific capacity, permeate, two-stage reverse osmosis plant,

1. INTRODUCTION

Nowadays the method of membrane desalination is widely used for processing mineralized water solutions, chemical and pharmaceutical liquid products, washing and working solutions, make-up water of machine-building, food, chemical, textile, fuel and energy and other branches of industry [1-5]. The traditional desalination method for such aqueous media includes pre-treatment and one- or twostages reverse osmosis desalination plants. Membrane desalination is also increasingly used in the treatment of solid waste landfills leachate. The treatment of highly concentrated waste leachate is a more complex multi-stage technological process [6-9]. It depends on the complex and unique composition of contaminated leachate, and the treatment process of such wastewater is studied in the differentiated manner and simultaneously with the search for the most appropriate integrated technologies, applying experimental installations if possible. But anyway, the stage of membrane desalination remains one of the key stages in the treatment of highly mineralized leachate. The final target of contaminated leachate desalination is to receive maximum amount of processed water meeting the discharge requirements into the fishery water bodies (permeate) and the minimum amount of concentrate to be returned to the landfill site. To reach this objective, it is necessary to develop membrane separation technological diagram, providing multi-stage treatment of landfill leachate, and reduction of both permeate and concentrate amount.

The research objective is to evaluate the membrane separation effectiveness for highly mineralized leachate applying pilot two-stage reverse osmosis plant.

2. RESEARCH OBJECTS AND METHODS

The experiments focused on deep desalination of preprepared leachate from the landfill "Dmitrovskiy" were conducted using two-stage reverse osmosis membrane pilot plant developed by "BMT ltd." in Vladimir, shown in Figure 1.



Figure 1: Two-stage reverse osmosis membrane pilot plant.

The principle technological diagram of pilot twostage reverse osmosis plant is shown in Figure 2.



Figure 2: Principle technological diagram of pilot two-stage reverse osmosis plant

SP1, SP2 - submersible pumps; C1, C2 - container; c1, c2, c3, c4, c5 - crane; V1,V2,V3,V4,V5 - valve; PG1, PG2,PG3, PG4 pressure gauge; F1, F2 - flowmeter; A1, A2 - membrane apparatus; S - sampler; P1, P2 - high pressure plunger pump

The clarified initial leachate after pre-treatment using granular filter was poured into the tank C1 and further was fed to the first stage of reverse osmosis membrane plant A1 by the high-pressure plunger pump P1. At the first stage of the plant under the operating pressure of up to 60 atm., the waste water was split into two streams: purified and partially desalinated water (first stage permeate), collected in the tank C2, and the first stage concentrate which was constantly looped to the tank C1. When reaching the specified degree of permeate collection, the process of waste water concentration was stopped. After the first stage of membrane separation concentrate from tank C1 was discharged into the intermediate auxiliary tank. The tank C1

was washed with tap water and permeate from the first stage of membrane separation was pumped into the tank C2. The first stage permeate was fed to the second stage A2 membrane apparatus by a plunger pump P2. The waste water was finally desalinated at the second stage under the operating pressure of max 15 atm.

The operating pressure at the inlet and outlet of the membrane vessels A1 and A2 was monitored according to the pressure gauges PG2 - PG4 and PG1 - PG3 readings respectively.

The permeate flow rate at stages 1 and 2 is determined by flow meter F1, and the concentrate flow rate - according to the flow meter F2.

Physical and chemical parameters detection in the feed waste water, as well as purified water after the first and second membrane filtration stages was carried out in compliance with GOST and Federal Environmental Regulations.

Since landfill leachate contains sufficiently large amount of mechanical and colloidal particles, as well as highly molecular organic substances, leachate samples were preliminary treated using laboratory mechanical granular filter with two-layer loading, consisting of anthracite and quartz sand, before studying the process of membrane filtration,

The membrane element RE 4040SN produced by CSM (South Korea) was chosen for the first stage of reverse osmosis separation for contaminated leachate and membrane element RE 4040BL was used for the second stage of reverse osmosis separation.

The initial solution composition of contaminated leachate is shown in table 1.

unit landfill leachate pH unit 7.91 PND F 14.1:2:34.121-97 Hardness mg·eqv/l 32 GOST 31954- 2012 Calcium mg/l 400 GOST 31869- 2012 Magnesium mg/l 144 GOST 31869- 2012	Item	Measure	Fed	Regulation
leachate pH unit 7.91 PND F 14.1:2:34.121-97 Hardness mg·eqv/l 32 GOST 31954- 2012 Calcium mg/l 400 GOST 31869- 2012 Magnesium mg/l 144 GOST 31869- 2012		unit	landfill	
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	Magnesium	mg/l	144	GOST 31869-
A11 1: : / /1 400 COST 21057		-		2012
Alkalinity mg·eqv/l 400 GOST-3195/-	Alkalinity	mg·eqv/l	400	GOST -31957-
2012				2012
Boron mg/l 17.2 GOST 31949-	Boron	mg/l	17.2	GOST 31949-
2012		-		2012
Iron (Fe _{total}) mg/l 12.49 PND F	Iron (Fe _{total})	mg/l	12.49	PND F
14.1:2:4.50-96				14.1:2:4.50-96
Cadmium mg/l 0.0003 GOST <u>31870</u> -	Cadmium	mg/l	0.0003	GOST <u>31870</u> -
2012				2012

Table 1: Initial solution composition of contaminated leachate

Silicon	mg/l	37.83	PND F
	_		14.1:2:4.215-06
Manganese	mg/l	1.36	GOST 4974-2014
Zinc	mg/l	0.18	GOST <u>31870</u> -
			2012
Ammonia	mg/l	6300	GOST 33045-
$(\mathrm{NH_4}^+)$			2014
Nitrates	mg/l	482	GOST 18826-73
(NO ₃)			
Sulphates	mg/l	50	GOST -31940-
(SO ₄)			2012
Chlorides	mg/l	7112	GOST 4245-72
(Cl ⁻)	_		
Anionic	mg/l	65	PND F
surfactants			14.1:2:4.15-95
Nonionic	mg/l	46.25	PND F
surfactants			14.1:2:4.194-2003
Petroleums	mg/l		GOST 31953-
	_		2012
COD	mgO ₂ /l	10733	GOST P 52708-
	_		2007
Color	degree	18000	GOST -31868-
	-		2012
Salinity	mg/l	31500	GOST P 54316-
-	-		2011

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3. RESEARCH RESULTS AND DISCUSSION

The chemical composition analysis of the initial contaminated leachate revealed its high alkalinity index. To reduce it, it was decided to make pH correction of the fed leachate solution in order to decrease calcium carbonate concentration, which easily precipitates on the membranes surface during concentrating. To assess the pre-acidification impact of the initial solution on first stage membrane separation, the experiments were conducted with preclarified leachate at of the initial sample solution pH and clarified filtration water acidified with sulfuric acid up to pH = 7. The process of first stage reverse osmosis separation was carried out at the pilot plant under the operating pressure of up to 60 atm., the initial solution feeding to the membrane element -1.3 m^3 /hour, the separated solution temperature - 25°C, the initial capacity of the membrane element for leachate - 225 l/hour. During the experiments, the dependence of membrane specific performance on the concentration degree (permeate selection) was determined without any pH adjusting of the feeding solution medium and with the initial solution acidification up to pH = 7. Test results are presented in tables 2 and 3.

Table 2: The dependence of the membrane specific performance on the concentration degree without initial solution pH adjusting.

Concentrat	t, °C	Element	Membrane	Permea
ing time,	,	capacity,	specific	te
min		l/h	capacity,	selectio
			$1/m^2 h$	n rate.
			-	%
0	14	225	32.1	-
5	14.5	159	22.7	5.7
15	15	133	18.9	14.3
35	20	106	15.1	28.5
50	25	62	8.9	42.9
60	26	58	8.3	45.7
65	30	46	6.5	57.1
	50	10	0.0	57.1
80	21	24	1.9	65 7
00	51	34	4.0	05.7
1				

Table 3: The dependence of membrane specific performance on concentration degree during acidification of the initial solution.

Concentr	t, °Č	Element	Membrane	Permeate
ating		capacity,	specific	selection
time,		l/h	capacity,	rate, %
min			$l/m^2 h$	
0	20	287	41	-
10	22	228	32.6	17.1
15	23	193	27.6	28.6
20	24	141	20.1	40.0
30	25	101	14.5	51,4
40	26	65	9.2	57.1
45	27	48	6,9	62.8
60	30	41	5.8	65.7

The comparative analysis of tables 2 and 3 shows that preacidification of the initial clarified leachate before reverse osmosis separation allows to increase membrane specific performance significantly, that means that the process of the first stage reverse osmosis separation is more efficient.

The chemical analysis of the first stage permeate (table 4) showed that the first stage reverse osmosis membrane treatment of the contaminated leachate allows to reaching sufficiently high degree of contaminants removal from the leachate, but permeate quality regarding a number of indicators (ammonia, nitrates, chlorides, total salinity) does not meet the standard parameters. In order to reach the required degree of purification, experiments on the first stage permeate of reverse osmosis separation were carried out, i.e. the second stage treatment of leachate was performed.

The second stage reverse osmosis separation was carried out under the operating pressure up to 15 bar, initial solution feeding to the membrane element of 1.3 m³/hour, the filtered solution temperature of 25 ° C, initial performance of the membrane element of 319 l/h.

In the course of experiments on reverse osmosis separation at the second stage of treatment, the dependence of the membrane specific performance fluctuations on concentration degree of (permeate selection) was also determined. It is shown in Figure 3.



Figure 3: Dependence of the specific productivity of the membrane on the degree of filtrate selection

The chemical analysis results of leachate reverse osmosis desalination after the first and second stages are presented in table 4.

	Reverse osmosis	
Item, unit	separation	
	1-st stage	2-nd stage
	permeate	permeate
pН	6.69	6.1
Hardness, mgeqv/l	0.18	0.04
Calcium, mg/l	2.6	0.6
Magnesium, mg/l	0.6	0.12
Alkalinity, mgeqv/l	20	3.2
Boron, mg/l	6.1	4.7
Iron (Fe _{total}), mg/l	0.01	0.01
Cadmium, mg/l	n/d	n/d
Silicon, mg/l	0.15	0.04
Manganese, mg/l	0.0016	0,004
Zinc, mg/l	0.001	n/d
Ammonia (NH ₄ ⁺), mg/l	276	54
Nitrates (NO ₃), mg/l	110	33
Sulphates, mg/l	18	9
Chlorides (Cl ⁻), mg/l	420	46
Anionic surfactants, mg/l	1.25	0.4

Table 4:	Chemical analysis results of leachate reverse osmosis
	desalination after the first and second stages

Item, unit	Reverse osmosis separation	
	1-st stage	2-nd stage
	permeate	permeate
Nonionic surfactants, mg/l	< 0.02	< 0.02
Petroleums, mg/l	0.25	0.12
COD, mgO ₂ /l	91	29
Color, degree	17	12
Salinity, mg/l	1675	232

Analyzing the research data, it can be concluded that twostage membrane reverse osmosis treatment of contaminated leachate provides the required degree purification rate thus meeting the discharge requirements for fishery water bodies regarding all indicators, except for ammonium nitrogen. To reach the standard indicator, the third stage of reverse osmosis separation is no longer reasonable, so the additional technological stage is required to process the second stage permeate by using other physical and chemical methods, such as adsorption or ion exchange.

4. CONCLUSION

Based on the research results we have worked out the optimum treatment parameters for pre-clarified concentrated leachate using pilot two-stage reverse osmosis plant: operating pressure at the 1-stage reverse osmosis was 60 kgf/cm², concentration degree K = 2.9; operating pressure at the 2-stage reverse osmosis was 15 kgf/cm², concentration degree K = 9.

The membrane specific performance showed the significant increase if initial solution is acidified and decreases alongside concentration increase.

The developed two-stage reverse osmosis pilot plant provides solving the problems of processing highly concentrated waste water not only from landfills, but also mineralized aqueous media of such industries as engineering, chemical, food, power engineering, petroleumrefining and others.

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