

Application of reverse osmosis to treat high ammonia concentrated reject water from sewage sludge digestion

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ABSTRACT

Reverse osmosis (RO) is well known as an efficient tool to remove different kinds of pollutants and is successfully applied to treat municipal or industrial wastewater. Present wastewater discharge regulations require more and more efficient removal of ammonia and phosphates. To increase the efficiency of their removal, new technologies should be applied. The application of RO for biologically treated wastewater can be considered as an alternative to modernization of the biological treatment. Conventional biological wastewater treatment plants (WWTPs) apply aerobic or anaerobic sewage sludge digestion. It causes problems with reject water (RW) produced during sewage sludge dewatering. RW is usually returned to the beginning of the treatment process which results in a decrease of treatment efficiency. This occurs mainly due to a high load of ammonia nitrogen in RW, the concentration of ammonia nitrogen reaches 2,000 mg N-NH₄⁺/L. The paper presents a novel approach to removing ammonia from RW using RO membrane techniques. The developed technique provides efficient treatment of RW, RO concentrate utilization and production of quality water used for technological purposes in WWTPs. Removed ammonia and other pollutants are added to sludge and are withdrawn together with dewatered sludge. The presented data show that application of RO techniques provides more economically reasonable and reliable solution of wastewater treatment and reuse than conventional biological tools. A flow diagram of the process that describes principles of RW treatment by RO and ammonia balance is presented. The research indicated that the use of RO in full scale in municipal and industrial WWTPs which utilizes anaerobic or aerobic sewage sludge digestion would result in a significant decrease of contamination load in RW.

Keywords: Wastewater treatment; Reverse osmosis; Sewage sludge digestion; Reject water; Ammonia nitrogen

1. Introduction

Biological treatment of municipal and industrial wastewater requires more and more efficient removal of nitrogen and phosphate to prevent eutrophication of surface water. To increase the efficiency of ammonia removal from sewage, different technologies are developed and applied. This, in turn, requires additional financial support for wastewater treatment plants (WWTPs) modernization [1–9]. Fig. 1 shows conventional biological WWTP flow diagram and all necessary processes used for sewage and sludge treatment. In this article, a novel approach to removing ammonia nitrogen and other pollutants using membrane reverse osmosis (RO) techniques is presented. RO is well known to be an efficient tool for removing different kinds of contaminants and is successfully applied to treat water and sewage [1,10,11]. Many RO installations are successfully used for final treatment of biologically treated wastewater and to reuse for technological

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purposes [10-13]. A flow diagram of RO post-treatment is shown in Fig. 2. The main disadvantage of applying RO in WWTP schemes is the existence of concentrate flows which cannot be discharged as they contain all removed pollutants including especially nitrogen and phosphorus. The novel approach presented in this article proposes a concentrate utilization technique that consists of increasing RO recovery up to 0.99. RO concentrate that contains removed ammonia and other pollutants is added to raw sludge and withdrawn together with dewatered sludge. Conventionally biological treatment of wastewater is connected with the production of excess sewage sludge as a by-product. Large installations use anaerobic sewage sludge digestion. The biggest advantage is the production of heat and electric energy from biogas. Meanwhile, during the final dewatering of digested sludge, highly concentrated reject water (RW) is produced. Concentration of ammonia in RW reaches 500-2,000 mg N–NH,⁺/L. Usually, it is returned to the main line of a WWTP without any separate treatment (Fig. 1). Extremely high concentrations of ammonia in wastewater require developing new efficient tools to reduce it [1,3–5]. To efficiently remove ammonia, a lot of improvements to biological reactors might be require which causes a certain increase of their volume

and need major financial investments (membrane bioreactor [MBR], augmentation reactors, etc.).

A novel approach is presented to remove ammonia nitrogen from RW using the membrane technique treatment with RO. RO is well known as an efficient tool for removing different kinds of pollutants and contaminants from wastewater and is successfully applied to improve quality of biologically treated wastewater [1,6–9]. Fig. 3 shows a flow diagram to treat RW from sewage sludge dewatering with RO membranes. The RO unit operates in circulation mode. The volume of reject is decreased by 4–10 times. RO concentrate is withdrawn together with the sludge. Membrane product can be added to the feed water or to the treated water, depending on ammonia concentration.

A new field of RO application suggests using RO to treat RW generated during sewage sludge stabilization in biological WWTPs. RO product can be used for technical purposes and the concentrate can be returned back to be mixed with sludge, thus increasing its nitrogen and salt content. Therefore, ammonia nitrogen is finally withdrawn together with dewatered sludge. An RO unit provides treatment of RW and an increase of ammonia in dewatered sludge. A schematic flow diagram of RW RO treatment and salt balance is presented in Fig. 3.



Fig. 1. Conventional wastewater treatment flow diagram.



Fig. 2. RO post-treatment of biologically treated wastewater.

A new approach was developed by the authors to increase RO recovery up to 0.99 value and to withdraw retentate together with sludge [12,13]. The proposed flow diagram of the process of RO treatment for wastewater reuse and RO concentrate utilization is shown in Fig. 4. The first stage membrane is used to treat wastewater and remove ammonia. Concentrate of the first stage membrane is further treated by the second stage membrane that is used to decrease concentrate flow to reach 0.99 recovery value. Product flow after second RO stage is mixed with the feed wastewater.

Experimental research was conducted to evaluate membrane efficiency for removing main wastewater pollutants such as: ammonia nitrogen, nitrate, biological oxygen demand (BOD, ppm) phosphate, etc. Experimental parameters of RO treatment make it possible to find optimum values of recovery and product flow as well as membrane characteristics (rejection, pressure) to provide efficient treatment and concentrate utilization. The main aim of the research was to develop relationships between ammonia and other pollutants removal and recovery using membranes. The other goals were determination of maximum recovery value-mass balance to evaluate characteristics of membrane units and economical comparison between MBR, augmentation reactors and RO post-treatment of biologically treated water.

2. Experiments description, materials and methods

The conducted experimental program was aimed at development of relationships between concentrations of different pollutants in RO product water vs. membrane unit recoveries. The developed relationships can be used for RO wastewater treatment unit design in order to determine required membrane type, its surface and recovery. Wastewater treatment by RO can be divided in two stages: stage 1 provides wastewater treatment and production of quality treated water; stage 2 provides increase of RO unit recovery up to 0.99 and higher to decrease concentrate flow. Product water of the second stage can be added to the feed water. The experimental program consisted of three experimental series.

- Series 1: To develop dependencies between membrane removal efficiencies and membrane product flow of main wastewater pollutants and membrane unit recovery. These results provide characteristics of membrane unit that ensures quality treated water in the first stage of membrane treatment.
- Series 2: Treatment of the first stage concentrate to reach recovery up to 0.99 and prognosis of product water and concentrate chemical content.





Fig. 4. A newly proposed solution to reuse biologically treated wastewater for boiler feed and steam production.

• Series 3: To develop dependencies between membrane removal efficiencies and membrane product flow and recovery during sludge dewatering retentate treatment. Experiments were conducted using two different laboratory membrane units to treat wastewater on the first and second stages. Flow diagrams of both units were similar.

A flow diagram of this experimental procedure is presented in Fig. 5. Feed water is pumped from feed water tank (1) into membrane module (3) using centrifugal pump (2). The working pressure value was 8 bar. In RO module, feed water stream was separated into two streams: product water and concentrate streams. Product water was forwarded to product tank (4) while concentrate was returned to the feed water tank (1). Feed water volume was decreased 10 times throughout test runs. In the first experimental series, feed water volume equaled 100 L and by the end of each test run concentrate volume equaled 10 L. Low-pressure RO membrane elements were used (model 4040 BLN) supplied by CSM (Korea). Membrane surface in 4040 modules was 10 m². In comparison, membrane elements with nanofiltration membranes (4040 90 NE) and high removal RO membranes (4040 BE) were tested. Cross flow value in test unit was 300 L/h. To reach high recovery value up to 0.99, operation of the second stage membrane test unit was performed. The flow diagram of the second stage test unit was the same as shown in Fig. 5. Concentrate from the tank (1) after the first series (10 L) was used in the second experimental series. The tank volume in the second stage membrane unit was 10 L. First stage concentrate (feed water) from tank (1) was pumped to membrane module. Small nanofiltration membrane modules (1812 standard) model were used. The membrane surface in the module was 0.5 m². Nanofiltration membranes

(1812 90 NE model) and low-pressure RO (1812 BLN) were used. These modules were manufactured and supplied by CSM, Korea. In the second stage unit, a small pump was used produced by C.C.K., model R0 900, was used. The cross flow was 50 L/h and working pressure was 7 bar.

Wastewater after secondary sedimentation tanks was taken from the wastewater treatment facilities.

Table 1 presents wastewater composition and some examples of product water and concentrate compositions measured at different recovery.

For the third experimental series, RW after sludge dewatering was used. Excess sewage sludge with whey and flotation sludge in a dairy WWTP was stabilized in an anaerobic digestion chamber and next dewatered with a centrifuge. The amount of RW after sludge dewatering measured in the dairy WWTP during the research period was up to 10% of the total amount of raw dairy sewage. The average concentrations of pollutants in RW that were used in laboratory experiments were: chemical oxygen demand (COD, ppm) – 1,830 mg O_2/L ; TOC – 155.3 mg/L; ammonia nitrogen – 1,537.6 mg N-NH₄⁺/L and total phosphorous – 137.1 mg P/L.

3. Discussion of the results

Fig. 6 shows the results of test unit operation – the influence of ammonia and other pollutants concentrations on recovery. The higher recovery is, the less is membrane rejection of dissolved pollutants. Recovery is defined as a ratio of product flow to the feed water flow. In our experiments recovery is calculated as a ratio between the water volume in tank 1 at a certain moment of experiment to the initial feed water volume in tank 1 in the beginning of the experiment.



Fig. 5. A schematic flow diagram of laboratory RO unit used in experiments. 1 – Feed water tank; 2 – pump; 3 – spiral wound membrane module; 4 – permeate tank; 5 – heat exchanger; 6 – pressure gauge; 7 – feed water flow meter; 8 – permeate flow meter; 9 – concentrate flow meter; 10 – by-pass adjusting valve; 11 – feed water adjusting valve; 12 – concentrate adjusting valve; 13 – cooling water adjusting valve; and 14 – sampler.

Table 1

Wastewater chemical composition and composition of product and concentrate at different recoveries

No.	Components	Wastewater after biological treatment	Low-pressure RO membrane				Nanofiltration membrane		Regenerations (permitted
			RO product (recovery 0.5)	RO concentrate (recovery 0.5)	RO product (recovery 0.9)	RO concentrate (recovery 0.9)	NF product (recovery 0.99)	NF concentrate (recovery 0.99)	discharge regulations)
1	pН	7.9	6.65	7.6	6.8	7.7	7.1	8.0	6.5–7.5
2	NH ₄ ⁺ , ppm	1.27	0.15	2.6	0.5	10.1	3.5	97	0.189
3	(PO ₄) ^{3–} , ppm	0.17	0.02	0.13	0.18	1.6	1.5	8.32	0.4
4	TOC, ppm	190	32	220	48	384	-	_	-
5	BOD, ppm	7.24	0.7	7.7	2.16	13.8	4.64	114.5	
6	(SO ₄) ^{2–} , ppm	23	0.12	29	0.67	226	5.9	218	
7	Cl⁻, ppm	266	29	416	411.4	3,942	795	5,822	
8	Oil, ppm	4.1	0.02	7.7	0.2	31.1	-	_	
9	Detergents,	1.8	0.05	2.5	0.25	10.2	-	-	
	ppm								
10	TDS, ppm	465	50	1,280	250	3,580	1,215	17,160	



Fig. 6. Concentration values of different pollutants in RO product water vs. recovery.

Fig. 7 shows results of membrane removal efficiencies (rejection values) calculation of different pollutants vs. recovery. To determine the required recovery value that provides efficient removal of all pollutants and ensures high product water quality, we suggested developing the obtained relationships (Fig. 7) to show dependence of specific concentration values $(C/C_{reg.})$ and recovery values, as shown in Fig. 8. A specific concentration value is defined as the ratio of C/C_{a} where *C* is the concentration of removed pollutant in initial feed water to $C_{reg.}$ – a value required by discharge regulations. When concentration of the pollutant in the product water reaches the regulation value, the value C/C_{ree} equals 1. Thus, the recommended recovery can be determined by the cross point of the curves yielding concentration vs. recovery curves and the line parallel to abscissa corresponding to C/C_{reg} =1. As low-pressure membranes rejection of different ions depends significantly on working pressure and while osmotic pressure increases with water salinity, a number of experiments were conducted to understand the influence of feed water salinity (total dissolved solids [TDS, ppm] value) on ammonia



Fig. 7. Removal efficiencies of different pollutants vs. recovery. Low-pressure RO membrane, BLN type (CSM, Korea).



Fig. 8. The $C/C_{reg.}$ value vs. recovery (*C* is the concentration values of pollutants, ppm; $C_{reg.}$ is the value, required by water discharge regulations).

rejection. Fig. 9 shows dependencies between ammonia rejection and recovery in different cases of wastewater TDS. An addition of sodium chloride to the feed water was performed. To predict the concentrations of ammonia in RO product water, the obtained data were presented as a relationship of membrane removal (R, %) and concentration factor K (Fig. 10). The concentration factor K is defined as a ratio of initial feed water volume in tank 1 (Fig. 5) at the beginning of the experiment and the volume of concentrate in the feed tank 1 at a certain moment of the experiment. The concentration factor K value is connected with the recovery value Rec. according to the following equation: Rec. = K - 1/K.

This approach enables us to present dependence of rejection vs. *K* as an exponential function:

$$R = c \times K^{b}$$

where *R* is the membrane ammonia removal, %; *K* is the concentration factor that is related with recovery (Rec.) by the following equation: K = (1/1 - Rec.); *b* is the power index that can be determined using empirically obtained dependencies: b = -0.000248 (TDS - 430) for wastewater with TDS value 600–1,500 ppm; b = -0.000115 (TDS + 220) for wastewater with TDS value 50–600 ppm; *c* is the empirical coefficient



Fig. 9. Influence of feed water TDS on ammonia removal efficiencies: dependencies between ammonia removal by low-pressure RO membrane on recovery for different feed water TDS values (1 – feed water TDS 770 ppm; 2 – feed water after addition of 3,000 ppm of NaCl; and 3 – feed water after addition of 6,000 ppm of NaCl).

value that can be determined from the following empirical dependencies: c = 0.00485 (2,337 – TDS), for wastewater with TDS value 600–1,500 ppm; c = 0.008 (3,000 – TDS), for wastewater with TDS value 50–600 ppm.

After product quality water is produced a problem of concentrate (retentate) handling and utilization must be faced. As it is shown in Fig. 8, usually the recovery value for wastewater treatment ranges between 0.8 and 0.9. As it was suggested above, concentrate flow can be treated (decreased) by introduction of an additional membrane step to increase recovery up to 0.99 value. In our experiments the initial amount of wastewater was 100 L. After the amount of concentrate in the tank 1 reached 10 L, the experiments were stopped and concentrate was moved to another test unit. A flow diagram of the second test unit was the same, as shown in Fig. 5. The volume of tank 1 was 10 L. A small gear pump and small spiral wound modules of 1812 standard were used with low pressure and nanofiltration membranes. As TDS value of circulating concentrate increases throughout test runs, membrane product flow constantly decreases and concentrations values of different pollutants in product water constantly increase. Fig. 11 shows dependencies between TDS, ammonia and phosphate concentrations increase in wastewater concentrate (Fig. 11(a)) and retentate after sludge dewatering (Fig. 11(b)).

Figs. 12-15 demonstrate reduction of specific product flow throughout test runs of wastewater treatment both on the first and on the second stages. Fig. 12 shows decrease of specific product flow of different membranes with feed water TDS growth. Fig. 13 demonstrates reduction of specific product flow vs. recovery during wastewater concentrate treatment on the second stage. Fig. 14 shows results of product flow measurements on both stages for different wastewater TDS values. TDS values were changed by addition of different amounts of sodium chloride to the feed water in tank 1 (Fig. 5). Fig. 14 shows dependencies between specific product flow rates of RO and NF membranes and recovery for different feed water TDS values. It is obvious that product flow dramatically decreases when recoveries reach 0.95-0.99 values. It seems reasonable to use nanofiltration membranes on the second stage to safe membrane costs. Fig. 15 shows dependencies between specific product flow throughout the whole process both for first and second stages and for different membranes applications.



Fig. 10. Ammonia removal vs. concentration factor *K* and feed water TDS (1 – TDS 300 ppm; 2 – TDS 500 ppm; 3 – TDS 750 ppm; and 4 – TDS 1,000 ppm).



Fig. 11. Ammonia and phosphate concentrations in the second stage product vs. recovery. (a) Treatment of wastewater retentate after the first stage treatment and (b) treatment of reject after digested sludge dewatering.



Fig. 12. Treatment of biologically treated wastewater at high recoveries. Dependencies between specific product flow rate and feed water TDS for different membrane types (1 – low-pressure RO, BLN type; 2 – nanofiltration membrane, 90 NE type and 3 – high removal membrane, BE-type). Membranes produced by CSM (Korea).



Fig. 13. Specific product flow rate on the second stage vs. recovery.



Fig. 14. Dependencies between specific product water flow rates and recovery for different feed water TDS values (1 - 300 ppm; 2 - 600 ppm and 3 - 1,000 ppm).



Fig. 15. Reduction of specific product flow of membranes vs. recovery throughout wastewater treatment at high recoveries using different membranes and concentrate utilization.

Figs. 10-15 demonstrate an experimental approach to determine required membrane characteristics to design a membrane unit for wastewater treatment and concentrate utilization. The main characteristics to be determined are: product quality, specific product flow and required recovery as well as membrane types on the first and second stages membrane units. Fig. 10 demonstrates an example to determine the required recovery value on the first stage. Ammonia concentration on the second stage for selected recovery can be determined using Fig. 11. The second stage product water can be added to the feed water. The second stage concentrate is added to sludge that is forwarded to a dewatering unit according to Fig. 2. The suggested improvement of conventional biological wastewater treatment includes the use of RO for wastewater post-treatment and the utilization of RO concentrate and retentate after sludge dewatering using additional RO step (Fig. 4). Fig. 16 shows a flow diagram of wastewater treatment and concentrate utilization as well as mass balance considerations to determine the required RO unit parameters to treat RW after sludge dewatering. The determination of required RO parameters and mass balance during treatment of RW after digested sludge dewatering is shown in Fig. 17.

When describing applications of RO tools to treat wastewater and RW from sewage sludge dewatering, it was



Fig. 16. Dissolved contaminants concentrations balance in the sludge sedimentation tank.



Fig. 17. Flow diagram and salt balance of reject water after digested sludge dewatering treatment by RO.

assumed that the feed water stream is adequately pretreated using ultrafiltration. Also, suspended colloidal, organic and bacterial fouling, as well as scaling can occur on membrane surface when high recoveries reached. A great deal of research work was undertaken by our team to investigate fouling and scaling processes [12,13]. As a result, "open channel" modules were developed that can be operated using feed water with high fouling and scaling potential without a hazard of decreasing membrane performance [12]. Even direct wastewater treatment without biological step was successfully carried out through the use of newly developed "open channel" modules [12]. The experiments conducted in this article provided no evidence of scaling and low soluble salts deposition from concentrate volume. The developed "open channel" RO modules [12] can be used to ensure safe operation of the RO unit at high recoveries. For higher concentrations of calcium in the feed wastewater, deposition of calcium carbonate can occur on membrane surface and in concentrate flow at high recoveries. Thus, a new technique is developed and proposed to withdraw excessive hardness from RO concentrate and reduce concentrate hardness and TDS at high recovery [13]. The presented data show that the application of RO techniques provides more economically reasonable and reliable solutions than biological tools. Future research will provide economical and technical survey and analysis to evaluate and compare modern methods to reduce ammonia and other biogenic elements in wastewater effluents.

4. Conclusions

Principles of the use of RO techniques in wastewater treatment schemes to improve product water quality are presented. RO concentrate which contains ammonia nitrogen and other pollutants is added to sludge and withdrawn together with dewatered sludge. The study showed high efficiency of RO in removing main pollutants from RW generated during anaerobic sewage sludge stabilization in a dairy WWTP. The research indicated that the use of RO in full scale in dairy WWTPs would result in a significant decrease of contamination load in RW. It might assure a stable and efficient functioning of dairy WWTPs without the necessity of biological stage modernization. Concentrate produced during RO treatment can be used for fertilizer production or blended with dewatered sludge before its final use.

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