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TREATMENT OF INDUSTRIAL WASTEWATER IN THE SEQUENTIAL MEMBRANE BIOREACTOR

OCZYSZCZANIE ŚCIEKÓW PRZEMYSŁOWYCH W SEKWENCYJNYM BIOREAKTORZE MEMBRANOWYM

Abstract: The aim of presented study which was associated with modification of the various work cycle phases duration in the membrane bioreactor, was to reduce the concentration of phosphate phosphorus during the leachate co-treatment with dairy wastewater. The experimental set-up was comprised of the membrane bioreactor equipped with the immersed membrane module installed inside the reactor chamber, and the equalization tank. During the co-treatment experiment performance the excessive activated sludge was constantly removed from the membrane bioreactor in order to keep its concentration at 3.5 g/dm³. The load of the sludge with the contaminants was equal to 0.06 g COD/g d.m. d. The concentration of oxygen was equal to 3 mg/dm³. The share of the leachates in the co-treated mixture was equal to 10% vol. The membrane bioreactor worked as the sequential biological reactor, in two cycles per day. Duration of each phase was equal as follows: filling - 10 min - with concurrent mixing phase lasting for 4 h, aeration phase - 1 h, sedimentation - 30 min and removal from purified wastewater - 30 min. After 4 weeks under these conditions, the modification of the sequential membrane bioreactor's work cycle was made. The duration of particular phases was shortened and two phases of denitrification and nitrification were introduced. Work cycle phases were modified as follows: filling - 10 min - with concurrent mixing phase lasting for 3 h, aeration phase - 4 h, mixing phase - 1 h, aeration phase - 3 h, sedimentation - 30 min and removal from purified wastewater - 30 min. Based on research, it was found that the change in membrane bioreactors' work cycle affects the effectiveness of treated mixture. It was found that the applied modification of phases of the cycle of the MSBR did not affect the concentration of organic compounds and the no significant changes in the concentration of ammonium and nitrate nitrogen in the effluent from the bioreactor were observed, however, the total nitrogen removal efficiency increased by 50%. Alteration of MSBR reactor particular phases duration caused reduction of concentration of P-PO₄³⁻ from 4.7 to 2.9 mg/dm³.

Keywords: dairy wastewater, landfill leachate, SBR, membrane bioreactor

Introduction

The leachate from municipal landfills is most frequently co-treated in municipal waste water treatment plants with municipal waste water, and is less frequently treated on-site. The treatment of landfill leachate as compared to the treatment of municipal waste water is much more challenging. This can be attributed to the fact that leachate contains high concentrations of organic substances (including toxic compounds), and that its composition

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and amount changes as the landfill ages [1, 2]. Currently, industrial waste water has to meet high quality requirements before it is discharged into surface water or into a municipal sewerage system. The permissible values of physico-chemical parameters characterizing the quality of leachate waters are regulated in the Regulation of the Minister of Construction of 14 July 2006 on the manner of performing the obligation of industrial sewage supplier and on conditions of discharging sewage to sewerage systems and the Regulation of the Minister of Environment of 18 November 2014 on conditions that have to be met when discharging sewage to water or to soil and on substances particularly hazardous to aquatic environment J. of Laws item. 1800 [3, 4]. In order to meet these requirements, especially when discharging the effluent into surface waters it is necessary to use highly efficient treatment systems, in which the removal efficiency of contaminants depends on the intended degree of treatment. In recent years, the possibility of treating landfill leachate in SBRs, which are an alternative to the conventional activated sludge method, has received considerable attention [5-11]. Sequencing batch reactors are successfully used for the treatment of municipal and industrial waste water and reject waters generated during the processing of sewage sludge. This technology offers feeding of the waste water into the reactor under alternating anaerobic-aerobic conditions during the filling phase and the use of internal recirculation of biomass. This ensures high removal efficiency of carbon, nitrogen and phosphorus while eliminating the phenomenon of sludge bulking. Thus, the use of a membrane bioreactor system operating as an SBR offers a number of advantages. The biological treatment of leachate in SBRs is preferable due to the fact that it has the ability to customize the system configuration and mode of operation for both short-term, daily and long-term changes in sludge loading. It is possible to introduce numerous technological modifications during the operation, such as modifying the duration of the operating phases or the operating cycle. It is also important that in comparison to the conventional activated sludge method the use of a secondary clarifier can be omitted, and thereby the footprint of the treatment system can be reduced. It is also possible to maintain high concentration of sludge in the membrane bioreactor, which corresponds to increased capacity of a treatment system [12-19].

Abood et al [8] treated the leachate from the municipal landfill in Chang Shankou (China) in the system comprising desorption of ammonia-coagulation-SBR-sorption on activated carbon [8]. In this study, each cycle of an SBR consisted of successive phases: filling (0.5 h), anaerobic phase (2 h), aerobic phase (8 h), anaerobic phase (2 h), sedimentation (1 h), and discharge of treated waste water (0.5 h). The efficiency of biological treatment of leachate was as follows: for COD 84.8%, for BOD₅ 88.4%, and for N-NH₃ 65.0%. Research on the treatment of landfill leachate in SBR reactors was also carried out by Uygur and Kargi [9]. The biological treatment of leachate was conducted in a 21-hour cycle of 3, 4 and 5 phases consisting of: anaerobic/anoxic/aerobic phases (2.0/1.0/4.0 h), anaerobic/aerobic/anoxic/aerobic phases (1.0/1.5/1.5/3.0 h) and anaerobic/anoxic/aerobic/anoxic/aerobic phases (1.0/1.0/2.0/1.0/2.0 h), respectively. The best results of treating landfill leachate were obtained in a five-phase system, namely COD of the treated waste water was 1400 mg/dm³, N-NH₄⁺ was equal to 107 mg/dm³, and P-PO₄³⁻ was 65 mg/dm³. In contrast, Laitinen et al [10] attempted to compare the efficiency of treating leachate from a municipal landfill in an SBR and in a batch-fed membrane bioreactor equipped with a Canadian hollow-fiber ZeeWeeD® immersed module. The authors showed that in both systems the removal efficiency of ammonium nitrogen was above 97.0%, while higher removal of BOD₇ (97.0%), phosphorus (88.0%) and total

nitrogen (50.0-60.0%) was observed in a membrane bioreactor. Also, the removal of suspended solids was higher in a membrane bioreactor than in an SBR [10].

In summary, the above-mentioned information suggests that the use of membrane bioreactors for the waste water treatment has become increasingly popular in recent years. They seem to be a good alternative to conventional treatment systems at high loading of organic and nutrient compounds, and characterized by high concentration of sludge.

Materials and methods

The subject of this study was leachate from a stabilized municipal landfill and waste water from the dairy industry. The dairy waste water originated from washing and rinsing processes, including washing of containers and devices, process equipment, floors and pipelines. The dairy waste water contains leaks and loss of milk from the production process and cleaning substances responsible for large load of phosphorus and pH fluctuations. The equipment and devices were washed with 30-60% sodium hydroxide, 15-30% nitric acid and 5-20% orthophosphoric acid. The dairy waste water mixed with 10% (by volume) of the leachate was subjected to biological co-treatment. The leachate and the dairy waste water were stored at 4°C. Average concentrations of individual pollutants characteristics of these waste waters are given Table 1.

Table 1
Characteristic of landfill leachates, dairy wastewaters and wastewaters co-treated in membrane bioreactor

Parameter	Unit	Leachate	Dairy waste water	Dairy waste water + 10 vol.% of leachate
COD		3055	13040	11200
BOD ₅		160	3800	3500
TN	[mg/dm ³]	690.0	68.0	104.0
N-NH ₄ ⁺		675.0	9.0	57.0
N-NO ₃ ⁻		2.9	45.0	41.0
P-PO ₄ ³⁻		28.1	36.8	39.0
pH	[-]	8.20	9.10	8.80
Conductivity	[mS/cm]	12.94	4.20	4.70

The co-treatment of the leachate with dairy waste water was carried out in a sequencing membrane bioreactor (MSBR) and the schematic diagram of the experimental system is shown in Figure 1.

The experimental system consisted of a buffer tank, a membrane bioreactor fitted with an internal hollow-fiber ultrafiltration module and the treated waste water storage tank. The volume of the reactor was 20 dm³. The experimental system was also equipped with a vacuum pump allowing for backwashing the hollow-fiber membranes. The reactor and the buffer tank were fitted with water level sensors and a multi-function meter that was used to monitor the conditions of the biological process (pH, oxygen concentration, waste water temperature). The entire system was fully automated.

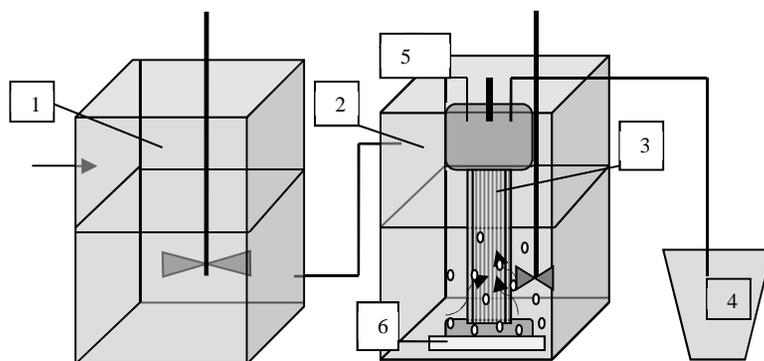


Fig. 1. The scheme of the experimental set-up: 1 - raw wastewater tank, 2 - aerobic-anaerobic chamber, 3 - capillary membrane module, 4 - purified wastewater tank, 5 - manometer, 6 - aeration

The biologically treated waste water was sucked into the fibers due to the vacuum in the membrane module and was pumped into the storage tank. The membranes were made of PES (polyether) with a cut-off of 80 kDa, and the filtration surface area was 0.4 m².

The study consisted in modifying the phases of the operation cycle of the membrane bioreactor, mainly due to the observed low removal of phosphorus. The operating cycle of the MSBR was modified by shortening the duration of the previous operating phases and by adding two more phases: anaerobic and aerobic phases. This modification was introduced to intensify the process of phosphorus removal from waste water, which required alternating aerobic-anaerobic conditions. In the preliminary part of the study the organisms present in the activated sludge were adapted to degrade the contaminants present in the co-treated waste water. The membrane bioreactor was operated in two cycles per day. In the first stage of the study the duration of the operating phases was: filling (10 min) with simultaneous mixing 4 h, aeration: 7 h, sedimentation and decantation of the treated waste water: 1 h. The modified operating cycle was as follows: filling (10 min) with simultaneous mixing 3 h, aeration: 4 h, mixing: 1 h, aeration: 3 h, sedimentation and decantation of the treated waste water: 1h. Excess sludge was regularly removed from the MSBR in order to maintain a constant concentration of 3.5 g/dm³. In the previous studies, excess sludge was removed from a reactor when its concentration was increasing, therefore the sludge retention time was approx. 20-30 d. Since the sludge was not removed in a continuous manner it was difficult to determine the exact sludge retention time. The study was conducted at a constant temperature of 20°C and the sludge loading was maintained at 0.06 g COD/g MLSS·d and the oxygen concentration remained at a level of 3 mg/dm³.

Next, due to exceeded concentration of total phosphorus in the effluent from the bioreactor it was attempted to reduce the sludge retention time and to determine its effect on the concentration of phosphate phosphorus in the treated waste water. In order to control the sludge retention time in the bioreactor once a day at the end of the aeration phase a calculated amount of mixed liquor was withdrawn from the reactor. At the beginning of the experiment the biomass concentration was equal to 4 g/dm³. Each time the sludge was withdrawn from the reactor the concentration of the activated sludge was decreasing until the sludge production was stabilized at 1.8 g/dm³.

The waste water treated under these conditions was polished in the reverse osmosis process. The process of pressure filtration was carried out in a dead-end system using

a GH-100-400 apparatus using a constant transmembrane pressure of 2 MPa and a rotational speed of the stirrer of 200 rpm. Osmotic membrane marked with the symbol SE was used. Table 2 shows the characteristics of this membrane. In order to determine the physical and chemical parameters of the leachate and the treated and untreated dairy waste water a set of analyses was carried out as shown in Table 3. Volumetric flux of the treated waste water was measured every day during the co-treatment of the leachate and the dairy waste water. In order to prevent a decrease in the permeability of the membranes the frequency of backwashing the fibers was determined.

Table 2

Characteristics of a flat membrane type SE

Polymer	Symbol	R [%]	pH [-]	ΔP [MPa]	Cl [ppm]	T [°C]
composite membrane with a thin layer of polyamide (TF)	SE	98.8	2-11	4	500	90

R - retention coefficient was determined for 1 wt.% solution of NaCl

Table 3

Physico-chemical determinations performed in the study

Parameter	Determination method
pH	Multi-function meter CPC 505 from ELMETRON
Conductivity	
COD	Test method - spectrophotometer MERCK PHARO 100 (measuring range: 10-150 mg/dm ³ , 100-1500 mg/dm ³ , 500-10000 mg/dm ³)
BOD ₅	Respirometric method using measuring system OXI Top WTW
Ammonia, nitrate, nitrite, total nitrogen	Test method - spectrophotometer MERCK PHARO 100 (measuring range: 2.0-75.0 mg NH ₄ -N/dm ³ , 5-150 mg NH ₄ -N/dm ³ , 0.2-20.0 mg N-NO ₃ /dm ³ , 0.03-2.30 mg N-NO ₂ /dm ³ , 0.5-15.0 mg N/dm ³ , 10-150 mg N/dm ³ , 0.5-30.0 mg P-PO ₄ /dm ³)
Phosphate phosphorus	

Results and discussion

Modification of phases in the operation cycle of the sequencing membrane bioreactor

In this study, in the first treatment step the influent contained significant amounts of nitrate and ammonium nitrogen, which could have hindered the removal of phosphorus in the anaerobic part of the cycle. Therefore, in order to increase the efficiency of phosphate removal it was decided to modify the operation cycle of the MSBR by changing the duration of the aerobic-anaerobic phases. Figure 2 shows the relationship between the concentration of organic compounds in the treated waste water of the based of modification work cycle phases in the sequential membrane bioreactor.

The introduced changes in the operation of the MSBR did not noticeably affect the concentration of organic compounds in the treated waste water. The values of COD and BOD₅ throughout the experiment remained at a constant level, which did not exceed the legal standards and were equal to 117 and 8 mg/dm³, respectively.

Other parameters of the waste water quality that were determined in this study were the nitrogen forms and the concentration of phosphate phosphorus. After the operation cycle of the membrane bioreactor was changed it was observed that the treatment efficiency of the investigated mixture of waste water improved. It was observed that the efficiency increased

with the time of operation. The obtained results are shown in Figures 3. The concentration of total nitrogen in the treated waste water at the beginning of the experiment was 14 mg/dm^3 and after the additional anaerobic-aerobic phases were introduced it decreased to approx 7 mg/dm^3 and remained at this level.

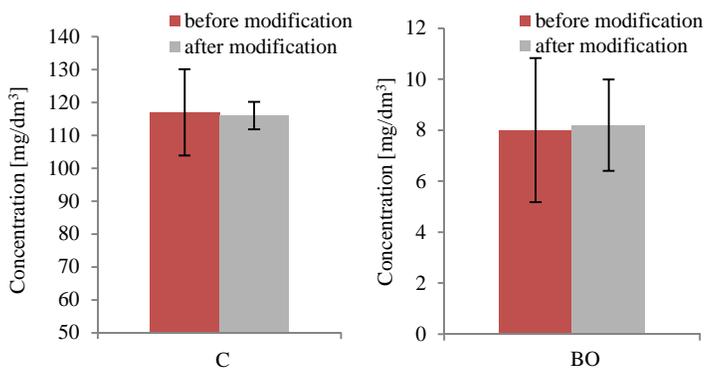


Fig. 2. Change of organic compounds concentration wastewater co-treatment the based of modification work cycle phases in the sequential membrane bioreactor (error bars show standard deviation of the mean for the obtained results, $n = 10$)

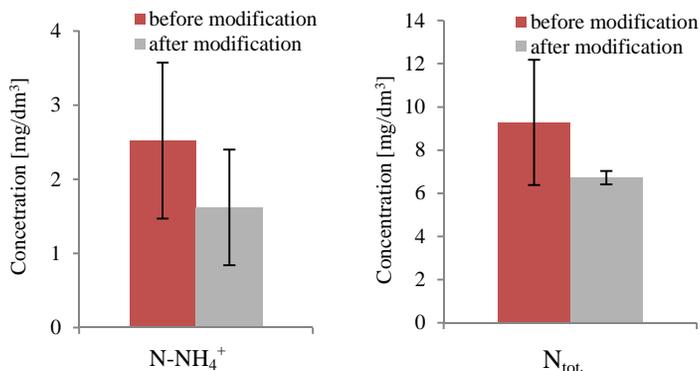


Fig. 3. Change of concentration total nitrogen and ammonia nitrogen of wastewater co-treatment the based of modification work cycle phases in the sequential membrane bioreactor (error bars show standard deviation of the mean for the obtained results, $n = 10$)

The ammonium nitrogen concentration was in the range of 1.0 to 3.9 mg/dm^3 throughout the entire experiment. Lower values of this parameter were observed after the modifications of the operation cycle of the sequencing membrane bioreactor. The concentration of nitrate nitrogen remained at the same level in both cases and varied in the range of 0.8 - 3.0 mg/dm^3 throughout the experiment (Fig. 4).

The changes in the operation of the MSBR were applied mainly due to excessive concentration of phosphate phosphorus in the effluent of the bioreactor, which was equal to 4.8 mg/dm^3 prior to the changes (Fig. 4). The modified duration of the cycle phases in the MSBR resulted in the decrease in the effluent concentration of P-PO_4^{3-} to the value of

2.9 mg/dm³, but this concentration also exceeded the permissible concentration specified in the Regulation of the Minister of Environment of 2009 (TP = 2 mg/dm³) [4]. In this case it was possible to use chemical precipitation of phosphorous as an additional unit process used periodically or in response to changing influent parameters. Chemical precipitation of phosphorus is widely used in municipal waste water treatment plants and serves as a method supplementary to biological processes [22].

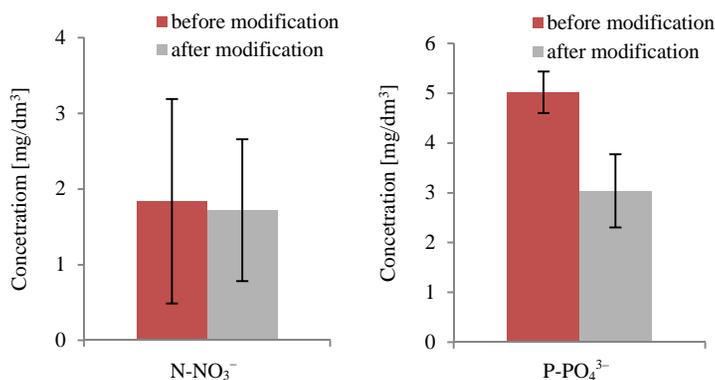


Fig. 4. Change of the concentration of phosphate phosphorus and nitrate nitrogen of wastewater co-treatment based on modification work cycle phases in the sequential membrane bioreactor (error bars show standard deviation of the mean for the obtained results, $n = 10$)

The effect of sludge retention time on the efficiency of industrial waste water co-treatment in the membrane bioreactor

As it is known, it is difficult in an SBR to achieve simultaneous removal of phosphorus and nitrogen. In general, the removal of phosphorus is acceptable at the short sludge retention time, which stimulates denitrification but hinders nitrification.

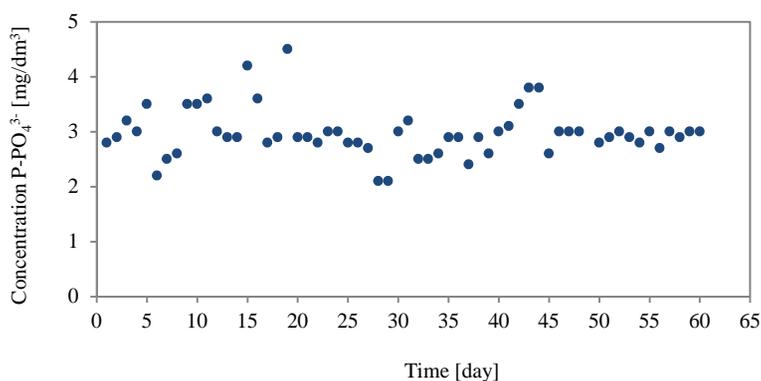


Fig. 5. Concentration of phosphate phosphorus in the co-treated waste water depending on to the time of the process at the shortened sludge retention time

Therefore, it was decided to shorten the sludge retention time to 10 d. Unfortunately, the concentration of phosphate phosphorus did not decrease in this system, and it was in the range of 2.1 and 4.5 mg/dm³. Also, the other parameters of waste water quality were equal to the values that were obtained for the most advantageous parameters of biological co-treatment of industrial waste water. The effect of shortening the sludge retention time on the concentration of phosphate phosphorus in the co-treated waste water is shown in Figure 5.

Reverse osmosis for the polishing of the waste water treated in the membrane bioreactor

Due to exceeded value of the concentration of phosphorus in the treated waste water it was decided to remove this nutrient by reverse osmosis as a polishing step. The volumetric flux of deionized water was $3.74 \cdot 10^{-6} \text{ m}^3/\text{m}^2\text{s}$ (2 MPa). The transport properties of the osmotic membrane are shown in Figure 6 and a change of the volumetric flux of polished waste water is shown in Figure 7.

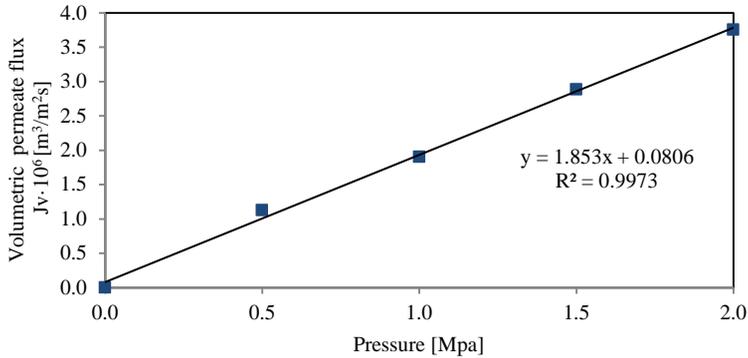


Fig. 6. Transport properties of the osmotic membrane

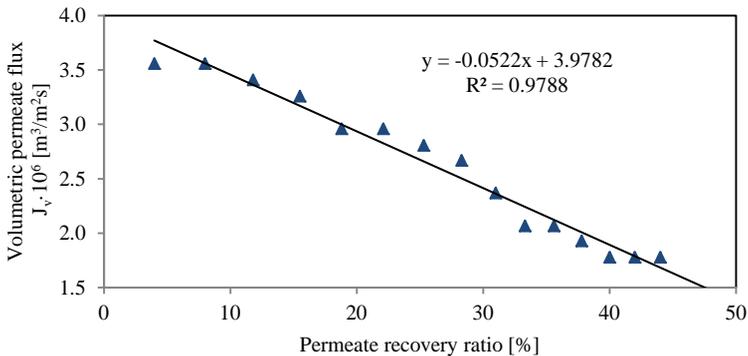


Fig. 7. The relationship between the volumetric flux of the treated waste water and the their recovery ratio in high-pressure membrane filtration

The permeate volumetric flux during the polishing of biologically treated waste water was being gradually reduced and after 50 vol.% of permeate was recovered it was equal to $1.48 \cdot 10^{-6} \text{ m}^3/\text{m}^2\text{s}$. The process of reverse osmosis allowed for effective polishing of waste water and for the removal of phosphorous compounds to the extent that the permissible values were not exceeded. The final concentration of phosphate ion was $0.5 \text{ mg}/\text{dm}^3$. Ammonium and nitrate ions were completely removed and the values of COD and TOC were $45 \text{ mg}/\text{dm}^3$ and $6.4 \text{ mg}/\text{dm}^3$, respectively.

The polishing of waste water in the process of reverse osmosis provided the opportunity to discharge it directly into the natural reservoir. The obtained results of the study are presented in Table 4.

Table 4
The efficiency of waste water co-treatment (5 vol.%) in the coupled system:
membrane bioreactor - reverse osmosis

Parameter	Dairy waste water + 10 vol.% leachate [mg/dm ³]	MBR effluent		RO effluent	
		Concentration [mg/dm ³]	Removal efficiency [%]	Concentration [mg/dm ³]	Removal efficiency [%]
COD	11400.0	115.0	98.8	45.0	60.0
BOD ₅	3500.0	8.0	99.7	0.0	100.0
TOC	825.0	24.0	97.1	6.4	73.5
TN	112.0	7.0	93.8	3.0	57.5
N-NH ₄ ⁺	59.0	1.5	97.5	0	100.0
P-PO ₄ ³⁻	46.0	3.0	93.4	0.5	83.3
N-NO ₃ ⁻	48.8	1.8	96.3	0.0	100.0
pH	8.2	8.7	-	7.1	-

Determination of the optimum backwashing frequency of hollow-fiber membranes

One of the main disadvantages of membrane bioreactors is that a filtration cake forms on the membrane surface, which leads to clogging of the pores. This is known as fouling and it results in a reduced flux of treated waste water during membrane filtration. In order to maintain a constant level of membrane permeability it is necessary to clean the membranes by physical or chemical methods. The most commonly used technique for cleaning the hollow-fiber module is backwashing the membranes with permeate or water (backflush method). In addition, fouling can be prevented when coarse-bubble aeration is used in a bioreactor tank. In order to prevent the decrease in the permeability of membranes backwashing of the fibers was applied in this study. The impact of this process on the flux of the treated waste water permeate is shown in Figure 8.

The deionized water flux determined for a non-operational membrane module was $11.8 \cdot 10^{-6} \text{ m}^3/\text{m}^2\text{s}$. The membrane bioreactor was also equipped with coarse-bubble aeration that was switched on periodically for 10 minutes at one hour intervals. After 7 days of operation of the membrane bioreactor under these conditions it was observed that the flux of treated waste water was reduced by 34% ($7.81 \cdot 10^{-6} \text{ m}^3/\text{m}^2\text{s}$). It was decided that the membrane backwashing would be performed every 7 days. After one-month of operation of the system before backwashing the membranes the volumetric permeate flux was $5.9 \cdot 10^{-6} \text{ m}^3/\text{m}^2\text{s}$. After each backwashing event the volumetric permeate flux was increasing in the range from 20 to 30%. Such a system provided a constant flux of treated waste water during the following months of operation and it was equal to $5.81 \cdot 10^{-6} \text{ m}^3/\text{m}^2$ after one year of the pressure filtration process being in operation.

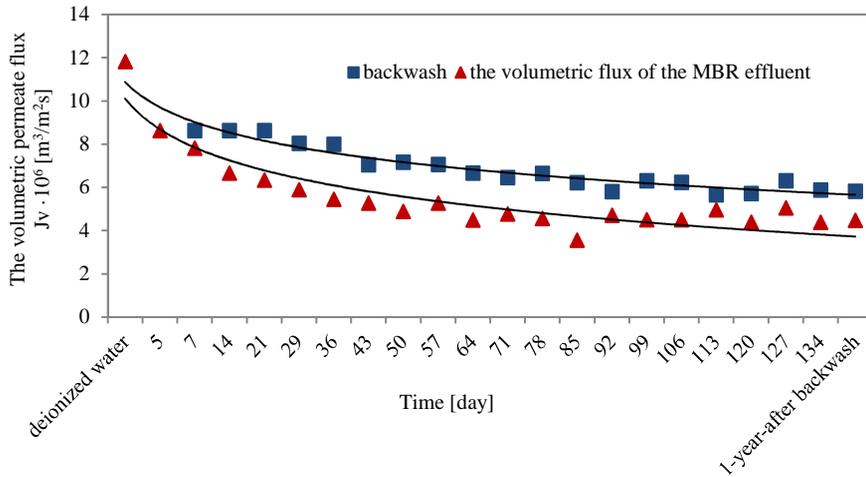


Fig. 8. Relationship between the volumetric flux of the MBR effluent and the time of the process and membrane backwashing

Conclusions

The following conclusions can be drawn from the present study:

1. It was found that the applied modification of phases of the cycle of the MSBR did not affect the concentration of organic compounds in the treated effluent.
2. No significant changes in the concentration of ammonium and nitrate nitrogen in the effluent from the bioreactor were observed, however, the total nitrogen removal efficiency increased by 50%.
3. The modified duration of the cycle phases in the MSBR resulted in a 40% decrease of effluent concentration of phosphate phosphorus, however, this concentration exceeded the permissible concentration specified in the Regulation of the Minister of Environment.
4. The shortened sludge retention time did not affect the removal efficiency of phosphate phosphorus removal.
5. The phosphate phosphorus concentration in the biologically co-treated waste water was 2.9 mg/dm^3 and it exceeded the permissible concentration. In order to increase the quality of the waste water to the level allowing direct discharge of into natural receiver, the waste water was polished by reverse osmosis.
6. It was found that the membranes should be backwashed at least once every seven days of the operation of a membrane bioreactor. Such a system provided a stable flux of treated waste water during the following months of operation and it was equal to $5.81 \cdot 10^{-6} \text{ m}^3/\text{m}^2 \cdot \text{s}$ after one year of the pressure filtration process being in operation.

Acknowledgements

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