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DETERMINATION OF THE MOST EFFECTIVE OPERATING CONDITIONS OF MEMBRANE BIOREACTOR USED TO INDUSTRIAL WASTEWATER TREATMENT

The effectiveness of co-treatment of leachates from an old site of municipal landfill with a dairy industry wastewater in a membrane bioreactor has been investigated. The subjects of the study were leachates collected at the municipal landfill in Tychy and dairy wastewaters from Dairy Plant in Bieruń. The determination of the most effective technological parameters of the process, i.e. activated sludge load and its concentration and types of COD fractions in the reactor influent was made. It was shown that the activated sludge load at the level of 0.06 g COD/(g_{DM}·d) was the highest permissible one among all the investigated loads and the process run effectively within the whole investigated range of concentrations of activated sludge in a bioreactor. It was found that in the treated wastewater, the fraction of easily degradable organic compounds was the dominant one.

1. INTRODUCTION

Due to the limited susceptibility of leachates components to biodegradation, novel technological systems are required assuring sufficient contaminants load removal from leachates before their deposition to natural collector [1–3]. Considering the fact that in Poland most of municipal landfills are stabilized ones (BOD_5/COD < 0.1) and the generated leachates contain significant amount of refractive substance, their co-treatment with dairy wastewater loaded with organic compounds of high concentration was examined. Post-production dairy wastewaters are generated during washing of milk transportation tanks and devices used to milk processing. They are mainly contaminated with organic substances like milk residues, whey and fats. They also contain washing agents responsible for the high load of phosphorus and pH variation [4–9].

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When the composition of dairy wastewater and leachates is known, a method of their co-treatment may be applied assuring both the sufficient removal rate of contaminants and direct deposition to the natural collector without influencing its water quality. A detailed characteristics of wastewater biodegradability in reference to usually applied COD/BOD_5 ratio can be determined by evaluation of COD fractions [9–11]. A basic balance of COD of particular fractions of organic compounds is given by the dependence [12, 13]:

$$\mathrm{COD} = S_S + S_I + X_S + X_I \tag{1}$$

where: S_S – COD of dissolved biodegradable compounds, S_I – COD of non-degradable dissolved substances, X_S – COD of hardly degradable organic suspensions, X_I – COD of non-degradable organic suspensions.

Wastewater treatment processes should provide high removal rate of refractive and nutrition (nitrogen and phosphorus) compounds as well as metabolites of microorganisms which participate in the treatment. Commonly used traditional, biological methods, e.g. activated sludge treatment are nowadays often modernized. The combination of biological process during which microbiological and biochemical decomposition reactions of organic contaminants occur with pressure-driven membrane filtration resulted in the introduction of a membrane bioreactor to the wastewater treatment technology. Such a solution has many advantages. One of them is the fact that the membrane bioreactor is never completely empty and the remained sludge participates in the next treatment cycle. It also favors the growth of microorganisms which already have been adapted to the effective process performance. The presence of membrane modules in the system practically eliminates necessity of the use of secondary settler tanks, assures longer retention time of hardly biodegradable high-molecular weight substances in the bioreactor and enables application of a higher concentration of activated sludge at low substrate load conditions [14–16].

2. MATERIALS AND METHODS

Apparatus. Biological co-treatment of leachates with dairy wastewater was carried out in a membrane bioreactor of the volume of 15 dm^3. The experimental set-up consisted of a membrane bioreactor equipped with an immersed membrane module installed inside the reactor chamber and the equalization tank (Fig. 1). Capillary membranes were made of polyvinylidene fluoride of the average pore size of 0.1 μm. The construction of the module enabled its back-washing performance. The back-washing was performed at various time-intervals, depending on the concentration of the activated sludge in the reactor and the rate of decreasing of the volumetric permeate flux. Additionally, at the bottom of the module along the capillaries the air forcing them to

wave and preventing the deposition of the biomass on their surface, was introduced twice a day.

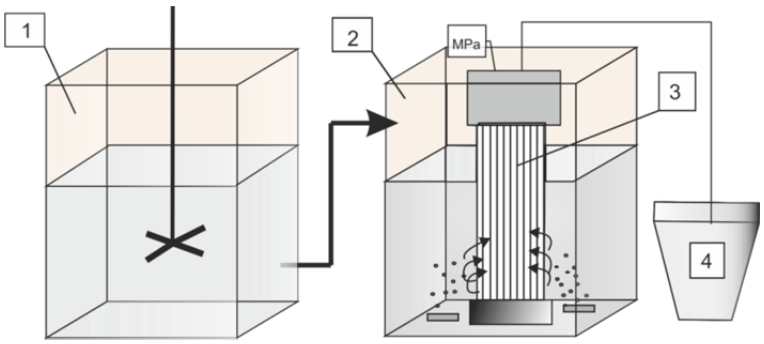

Fig. 1. Scheme of the experimental set-up: raw wastewater tank (1), aerobic-anaerobic chamber (2), capillary membrane module (3), purified wastewater tank (4)

Substrate of the study. Leachates generated at municipal landfill in Tychy--Urbanowice and dairy wastewaters from District Dairy Cooperative in Bieruń were investigated. They were stored at 4 °C. During the studies, the sampling of raw and purified wastewaters and evaluation of their quality were performed according to the Directive of The Minister of the Environment (DME) [17].

Table 1

Characteristics of landfill leachates, dairy wastewaters and wastewaters co-treated in the membrane bioreactor

Parameter	Leachates	Dairy wastewaters	Dairy wastewaters + 5 vol. % leachates
COD, mg O_2/dm^3	3040	2933–7030	3015–7010
BOD_5, mg O_2/dm^3	250	1200–2200	1200–2200
BOD_5/COD	0.08	0.3-0.5	0.3–0.5
TC, mg C/dm^3	1554	353–1438	402–1489
TOC, mg C/dm^3	354	295–1396	321–1359
$N\text{-}NH_4^+$, mg $N\text{-}NH_4^+$/dm^3	960	7–25	60–89
$N\text{-}NO_3^-$, mg $N\text{-}NO_3^-$/dm^3	4.1	7.8–20.9	7.4–20.5
$P\text{-}PO_4^{3-}$, mg $P\text{-}PO_4^{3-}$/dm^3	10.1	11.9–45.9	11.2–45.4
pH	7.6	6.5–8.4	7.0–8.4

It was found that the values of contaminants indicators which characterized particular samples of dairy wastewaters differed from each other, probably due to the

technological process carried on during their sampling. Part of samples originated directly from the production while the rest was collected from tanks and milk processing devices. Some physicochemical parameters of the landfill leachates and dairy wastewaters are presented in Table 1.

Method of the study. The studies were conducted in three stages. The former one was the adaptation of the activated sludge to treat leachates mixed with dairy wastewater in the membrane bioreactor. The 20 day old activated sludge collected at Wastewater Treatment Plant in Gliwice was used. The share of the leachates in the co-treated mixture was equal to 5 vol. %. After four weeks of operation, the activated sludge was adapted and reached the equilibrium state what was confirmed by the more or less constant values of contaminants indicators measured in the purified wastewater. During the co-treatment experiment, the excessive activated sludge was constantly removed from the membrane bioreactor in order to keep its concentration at the level of 3.5 g/dm^3. The load of the sludge with the contaminants was equal to 0.05g COD/(g_{DM}·d), and the concentration of oxygen was at the level of 3 mg/dm^3. The system was operated as the sequential biological reactor in two cycles per day. The duration of particular operation stages was as follows: filling and mixing phase – 4 h, aeration phase – 7 h, sedimentation and purified wastewater removal – 1 h. The next stage of the study was determination of the most effective activated sludge load with substrate which varied in the range of 0.05–0.1 g COD/(g_{DM}·d). The process was carried out at the same operating conditions as the ones applied during the sludge adaptation. In the final stage of the study, evaluation of the membrane module operation and the effectiveness of the treatment of dairy wastewater and landfill leachates mixture at the activated sludge concentration ranging from 4–16 g/dm^3 was made. At this stage, the load of the sludge with contaminants was kept constant at the level of 0.06 g COD/(g_{DM}·d).

Analytical methods. The evaluation of the effectiveness of the treatment process was based on the change of parameters characterizing the crude sewage and treated sewage. The following parameters were monitored: pH, COD, BOD_5, TOC, concentration N_{tot}, $N\text{-}NO_3^-$, $N\text{-}NH_4^+$, $P\text{-}PO_4^{3-}$, The oxygen concentration was measured using oxygen meter CO-411. The carbon analyzer Multi N/C (Jena Analytik) was used to determine particular forms of carbon. Total, nitrate and ammonium nitrogen as well as COD and phosphate phosphorus were measured according to the methods given by Merck company. The BOD_5 was determined by the respirometric method using OXI Top WTW analyzing set. COD fractions: S_S, S_I, X_S, X_I, were calculated based on the ATV-A 131 directive [12]. The method of determination of fractions was based on the measurements of COD and BOD_5 in filtered and non-filtered samples of purified and raw wastewaters [12, 18].

3. RESULTS AND DISCUSSION

3.1. DETERMINATION OF COD FRACTIONS IN A MIXTURE OF LANDFILL LEACHATE AND DAIRY WASTEWATER

The percentages of particular COD fractions in the co-treated wastewater is shown in Fig. 2. The analyzed samples of raw non-filtered co-treated wastewater characterized with the COD value at the level of 3030 mg/dm^3, 4730 mg/dm^3 and 3680 mg/dm^3, while BOD_5 values were equal 1500 mg/dm^3, 2200 mg/dm^3 and 1700 mg/dm^3, respectively.

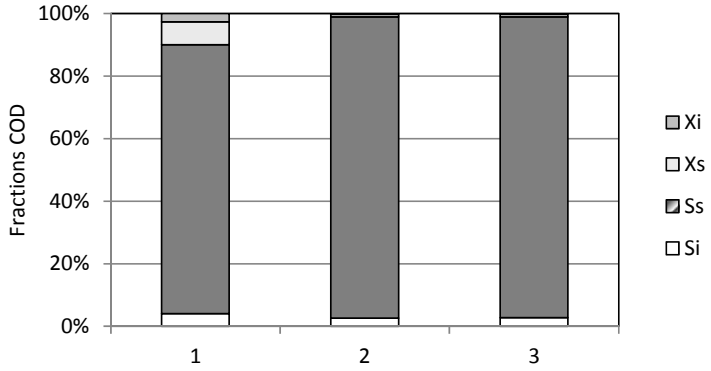

Fig. 2. The percentage share of particular COD fractions in the co-treated wastewater: 1, 2, 3 – dairy wastewater sampled within 3 months with the addition of 5 vol. % of municipal landfill leachates

The same indicators determined for purified wastewater were comparable and equal to 112 mg COD/dm^3 and 6 mg BOD_5/dm^3. Particular COD fractions in raw wastewater were as follows: fraction of dissolved biodegradable compounds *Ss* – 86–96.3%, while the fraction of hardly degradable organic suspensions X_S – 0.8–7.3%. The amount of non-degradable dissolved substances S_I varied from 2.6% to 4%, while in the suspension X_I they were in the range of 0.3–2.7%. The fraction of organic biologically degradable compounds in the co-treated wastewater was the dominant one, what confirmed the assumption that raw wastewater mixture was easily biodegradable. The comparison of the obtained results with those presented in [9–13] in which COD fractions were determined for municipal wastewater revealed that the contents of biologically degradable fractions (S_S and X_S) were lower than those determined for dairy wastewater.

3.2. THE DETERMINATION OF THE MOST EFFECTIVE ACTIVATED SLUDGE LOAD IN THE MEMBRANE BIOREACTOR

The mixture of raw wastewater which was introduced to the membrane bioreactor characterized with the high COD value in the range of 3500–7000 mg/dm^3. The effi-

ciency of decrease of the indicator value in the case of purified wastewater was high within the whole range of investigated loads and varied from 95.9 to 97.1%. It was shown that COD concentration in the bioreactor effluent increased with the increase of the activated sludge load with the contaminants (Fig. 3).

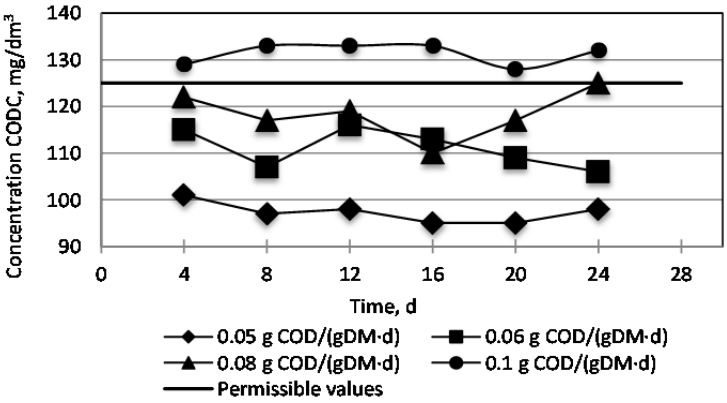

Fig. 3. Dependence of the COD concentration in the purified wastewater on the activated sludge load with contaminants and duration of the process performance in the membrane bioreactor

The average value of COD at the load of 0.05 g COD/(g_{DM}·d) was equal to 95 mg/dm^3, while for 0.06 g COD/(g_{DM}·d) it was 116 mg/dm^3. It was found that at the load of 0.08 g COD/(g_{DM}·d), the value of COD almost reached the permissible limit equal to 125 mg/dm^3. Only in the case of the highest sludge load, i.e. 0.1 g COD/(g_{DM}·d), the permissible value according to the Directive of The Minister of the Environment (DME) [17] was exceeded by 8 mg/dm^3.

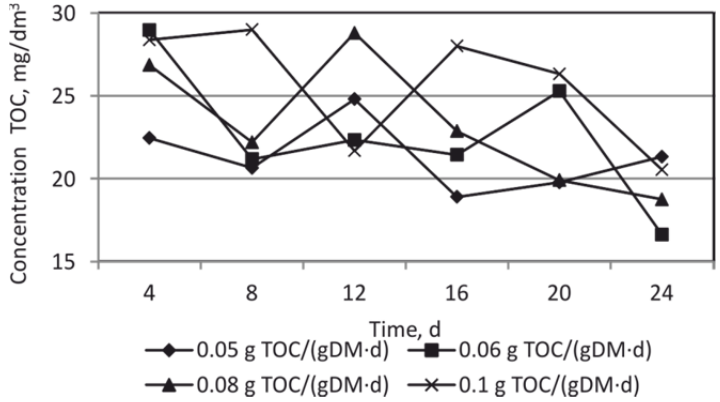

Fig. 4. Dependence of TOC content in purified wastewater on the activated sludge load with contaminants and the duration of the process performed in the membrane bioreactor

Another parameter characterizing purified wastewater was TOC. The dependence of total organic carbon on the activated sludge load with organic contaminants and the duration of the process is shown in Fig. 4.

The concentration of TOC determined for all investigated activated sludge loads did not exceed the permissible value according to DME (30 mg/dm^3). The observed slight differences, however, enabled determination of the most effective activated sludge load.

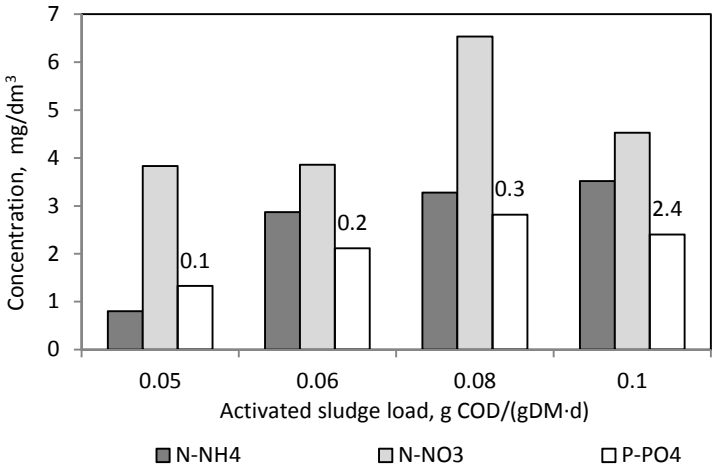

Fig. 5. Dependence of the concentration of nutrition compounds in the purified wastewater on the activated sludge load with contaminants

The concentration of nutrients in the treated wastewaters also slightly varied during the process. In Figure 5, the concentrations of $N\text{-}NH_3^+$, $N\text{-}NO_3^-$, $P\text{-}PO_4^{3-}$ ions in function of the applied activated sludge load is presented.

The efficiency of removal of ammonium nitrogen was high for the lowest applied sludge load i.e. 0.05 g COD/(g_{DM}·d) and it was equal to 98%, corresponding to the contaminant concentration at the level of 0.8 mg $N\text{-}NH_4^+/dm^3$. In the case of other applied loads, the concentration of the ion changed in the range of 2.9–3.5 mg $N\text{-}NH_4^+/dm^3$ and never exceeded the permissible value (10 mg $N\text{-}NH_4^+/dm^3$). The concentration of nitrate nitrogen measured in the mixture of dairy wastewater and leachates was equal to 4.8 mg $N\text{-}NO_3^-/dm^3$, while in the purified wastewater it was in the range of 3.8–6.5 mg $N\text{-}NO_3^-/dm^3$ depending on the sludge load (the permissible value – 30 mg $N\text{-}NO_3^-/dm^3$). It was shown the permissible concentration of phosphate phosphorus (2 mg P_{tot}/dm^3) was always exceeded regardless the sludge load, ranging from 1.3 to 3.1 mg $P\text{-}PO_4^{3-}/dm^3$. In the studies, $P\text{-}PO_4^{3-}$ was determined. According to

the literature PO_4^{3-} is about 50–70% of total phosphorus [19]. The lowest concentration of phosphate phosphorus (1.3 mg $P\text{-}PO_4^{3-}/dm^3$) was noted for the lowest activated sludge load, i.e. 0.05 g COD/(g_{DM}·d).

3.3. IMPACT OF THE ACTIVATED SLUDGE CONCENTRATION ON THE EFFECTIVENESS OF CO-TREATMENT OF LEACHATES WITH DAIRY WASTEWATER

The final stage of the study was the evaluation of the membrane module operation and the effectiveness of the wastewater treatment at the increasing concentration of dry mass of activated sludge. It was noticed that the increase of the activated sludge concentration did not affect the effectiveness of the co-treatment of municipal landfill leachates. The dependence of COD and TOC values measured in the purified wastewater on the activated sludge concentration in the membrane bioreactor is shown in Fig. 6.

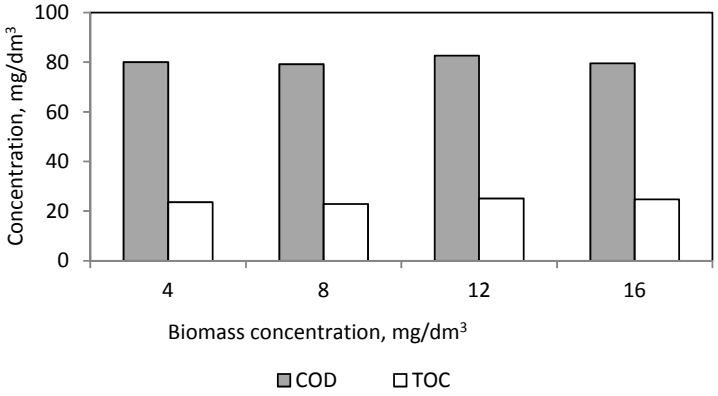

Fig. 6. Dependence of COD and TOC values characterizing purified wastewater on the activated sludge concentration

The analysis of the content of organic compounds in the purified wastewater revealed the high effectiveness of the treatment for all activated sludge concentrations. Both, COD and TOC values measured for every investigated concentration were at such a level that the purified stream could be directly disposed to the natural receiver. The COD concentration was in the range of 79–83 mg/dm^3, while TOC of 22–25 mg/dm^3. Next, the change of concentration of nutrition compounds i.e. ammonium nitrogen, nitrate nitrogen and phosphate phosphorus in dependence of the concentration of the activated sludge was analyzed, and the obtained results are presented in Fig. 7.

Regardless the applied activated sludge concentration, the concentration of phosphate phosphorus in the membrane bioreactor effluent always exceeded the permissible level. According to DME, the concentration of total phosphorus should not exceed

2 mg/dm^3. The values obtained during the experiments were equal in average to 3.7 mg/dm^3, 4.6 mg/dm^3 and 5.2 mg/dm^3 for the activated sludge concentrations 4 g/dm^3, 8 g/dm^3, 12 g/dm^3, respectively, while for the highest its concentration, i.e. 16 g/dm^3 it was equal to 4.8 mg/dm^3.

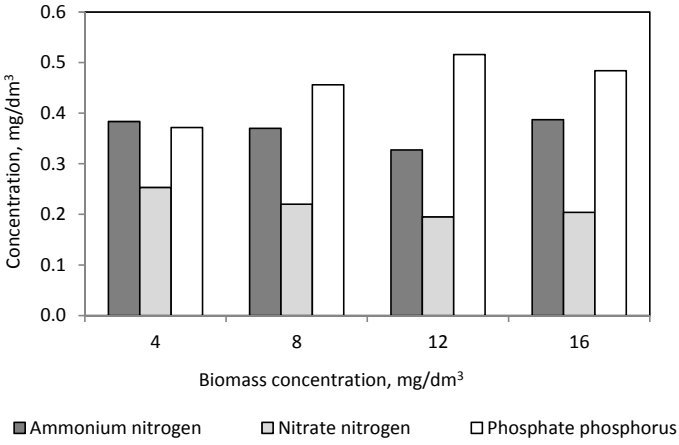

Fig. 7. Dependence of the concentration of nutrition compounds on the concentration of activated sludge

During the first stage of SMBR operation, the inflowing wastewater contained significant amounts of ammonium and nitrate nitrogen, what could have affected the dephosphatation in the anaerobic part of the cycle. Generally, phosphorus is effectively removed at young activated sludge age which also favours the intensive denitrification but it affects the nitrification process. Probably, in order to increase the removal rate of phosphates, the bioreactor operation should be modified, changing the duration of aerobic-anaerobic phases. In the case of other nutrition compounds, the effectiveness of leachates co-treatment was high for all investigated activated sludge concentrations. The removal efficiency of ammonium nitrogen was kept at the high level equal in average to 94%, corresponding to its concentration of 3.3–3.9 mg/dm^3.

A slightly higher removal rates were obtained for nitrate nitrogen. Its concentration in the bioreactor effluent was in the range of 1.7–2.5 mg/dm^3. The highest removal efficiency equal to 64.6% was obtained for the highest investigated activated sludge concentrations, i.e. 12 and 16 g/dm^3, while for the lowest one – 4 g/dm^3, 48% reduction was obtained.

During the study, the monitoring of the volumetric permeate flux of the purified wastewater was made for every activated sludge concentration in the membrane bioreactor. In order to prevent the blockage of the capillary membrane pores, the decrease of the volumetric permeate flux by 30% caused the application of the membrane backflushing with the permeate. It was found that the increase of the concentration of biomass in the reactor chamber had a significant influence on the module capacity. When

the concentration of the activated sludge was equal to 4 g/dm^3, the volumetric permeate flux was at the level of 8.8×10^{-6}m^3/(m^2·s), while at the concentration of 16 g/dm^3 it decreased to 5.6×10^{-6} m^3/(m^2·s). It can be supposed that at the proper control of the membrane module operation the increase of the activated sludge concentration in the reactor chamber would be is possible. It also enables the treatment of higher amount of wastewater and the operation at the lower activated sludge load as well.

4. CONCLUSIONS

COD fractions in the mixture of treated wastewater were as follows: biologically degradable fractions S_S and X_S from 93.6% to 96.3%, dissolved non-degradable organic substances S_I from 2.6% to 4%, and X_I from 0.3% to 2.7% in the suspension. The domination of the fraction of easily biodegradable organic compounds confirmed the high biodegradability of the treated wastewater.

The activated sludge load at the level of 0.06 g COD/(g_{DM}·d) turned out to be the highest permissible one to be applied during 2 cycles/day reactor operation. The purified wastewater characterized with the following values of parameters (mg/dm^3): COD – 113, TOC – 22.6, N-NH_4^+ – 2.9, N-NO_3^- – 3.9, P-PO_4^{3-} – 2.1.

The high effectiveness of wastewater treatment at the activated sludge concentration ranging from 4 g/dm^3 to 16 g/dm^3 was obtained. Both COD and TOC concentrations measured for all investigated activated sludge concentrations were at such levels, that the purified wastewater could be directly removed to the natural receiver. In the membrane bioreactor effluent, the COD concentration was equal to 79–83 mg/dm^3, while TOC – 22–25 mg/dm^3.

The concentrations of phosphate phosphorus in the purified wastewater exceeded the permissible value for all investigated activated sludge concentrations. Its concentration in the reactor effluent was in the range of 3.7–5.2 mg/dm^3.

The increase of the biomass concentration diminished the membrane permeability. The volumetric permeate flux at the activated sludge concentration 4 g/dm^3 was equal to 8.8×10^{-6}m^3/m^{2}s, while for 16 g/dm^3 it decreased to 5.6×10^{-6} m^3/m^{2}s.

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