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ALEKSANDRA PETROVIČ
MARJANA SIMONIČ

Faculty of Chemistry and Chemical Engineering, University of Maribor, Maribor, Slovenia

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THE EFFICIENCY OF A MEMBRANE BIOREACTOR IN DRINKING WATER DENITRIFICATION

Article Highlights

- High nitrate removal efficiencies (more than 90%) were obtained at flow rates below 4.8 L/h
- The denitrification efficiencies were highly dependent on the the hydraulic retention time
- The highest specific denitrification rate was achieved at 0.2738 g/L NO_3/(g/L MLSS d)
- The maximum reactor removal capacity was calculated at 8.7472 g NO_3/m^3 h

Abstract

The membrane bioreactor (MBR) system was investigated regarding its nitrate removal capacity from drinking water. The performance of a pilot-scale MBR was tested, depending on the operational parameters, using sucrose as a carbon source. Drinking water from the source was introduced into the reactor in order to study the influence of flow rate on the nitrate removal and denitrification efficiency of drinking water. The content of the nitrate was around 70 mg/L and the C/N ratio was 3:1. Nitrate removal efficiencies above 90% were obtained by flow rates lower than 4.8 L/h. The specific denitrification rates varied between 0.02 and 0.16 g/L NO_3*/(g/L MLSS d). The efficiencies and nitrate removal were noticeably affected by the flow rate and hydraulic retention times. At the maximum flow rate of 10.2 L/h still 68% of the nitrate had been removed, while the highest specific denitrification rate was achieved at 0.2738 g/L* NO_3*/(g/L MLSS d). The maximum reactor removal capacity was calculated at 8.75 g* NO_3/m^3 *h.*

Keywords: capacity, denitrification, drinking water, efficiency, membrane bioreactor, sucrose.

Drinking water sources contaminated by nitrate and nitrite represent one of the greater environmental concerns and risks for human health. High standards for drinking water are recommended by the Council of European Communities, a maximum of 11.3 mgNO_3-N/L for nitrate, and 0.03 mgNO_2-N/L for nitrite [1,2]. If denitrification efficiency is studied, accurate standard methods must be chosen in order to gain reliable analyses of water samples [3-6]. Several methods (physical, chemical, physico-chemical, and biological) for drinking water treatment have been examined to date. In the process of biological denitrification, which is considered as one of the more cost-effective and friendly methods in relation to the environment [7,8], bacteria use nitrate as an electron acceptor for respiration under anoxic conditions [1,9]. An external carbon source, *i.e.*, an electron donor, should be ensured for the activities of microorganisms during heterotrophic denitrification [10]. The presence of certain carbon sources importantly effects the denitrification rate and *COD* demand. The efficiency of heterotrophic denitrification is usually relatively high; nevertheless, the nitrate and nitrite accumulation have been found to inhibit complete nitrate removal [10-12].

Membrane processes could efficiently remove several contaminants from the contaminated water: organic and inorganic matter, bacteria, various suspended solids and residue carbon sources [13]. The membrane bioreactor (MBR) is a combined system of biological treatment and membrane filtration that offers several advantages: high effluent quality, low

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Correspondence: A. Petrovič, Faculty of Chemistry and Chemical Engineering, University of Maribor, Smetanova 17, SI-2000 Maribor, Slovenia.
E-mail: aleksandra.petrovic@um.si
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sludge production, lower needs of chemical agents, and high denitrification rates [14-16].

On the other hand, one of the major problems associated with conventional denitrification systems and MBRs is the problem of the direct contact between the denitrifying organisms and the water [17]. An interesting alternative for solving this problem is the membrane bioreactor/membrane contactor system (MBR/MC). In contrast with other MBR systems, this system uses a special membrane for separating the biomass from the water and therefore reducing the risk of induced microbial contamination [18]. A bench-scale microporous membrane bioreactor has also been developed for this purpose [17]. However, in conventional processess for drinking water treatment a process of disinfection is usually required that could increase the operational costs. Despite the above mentioned weaknesses the potential of a membrane bioreactor for nitrate removal in drinking water has been investigated by many other researchers [2,15,19-21].

The process highly depends on the operating conditions, particularly the inflow nitrate concentration and the flow rate. Other important factors in the removal of pollutants by the MBR system are the hydraulic retention and sludge retention times [15,22]. However, due to the formation of filter cake, membrane separation efficiency decays over time [23]. Recently, studies concerning membrane bioreactors and denitrification have mostly been related to the operation parameters such as pH and oxygen levels [12,24], as well as the different types and effects of external carbon sources [10,19]. Moreover, some investigations have focused on the kinetic properties of the process [1,9,12], and some on the modelling of the process [1,14,16]. As the MBR operation is associated with energy consumption and operational costs, many researches have examined the fouling itself and the factors influencing membrane fouling [16,25]. However, applications of MBRs in drinking water treatment have been very limited regarding nitrate removal capacity calculations.

The basic purpose of this research was to perform a series of experiments in order to study the efficiency of nitrate removal from drinking water. Denitrification is well documented in the literature, yet the novelty of present paper is in high process efficiencies using cost effective sucrose in comparison with more expensive carbon sources, such as ethanol, methanol, glucose, etc. The influences of flow rates and hydraulic retention time on the biological process in MBR were studied. The specific denitrification rate and the nitrate removal capacity of the reactor were calculated.

EXPERIMENTAL

Pilot plant and operational conditions

Drinking water enriched with nitrates, with sucrose as the carbon source, was introduced into a Zenon ZeeWed 10 membrane bioreactor. A series of experiments were carried out in order to study the efficiency of heterotrophic denitrification from drinking water samples. The inflow rates gradually increased from 0.6 up to 10.2 L/h. The steady-state conditions were ensured at each inflow rate. A submerged hollow-fibre membrane with a pore size of 0.04 μm was used in the experiments. The membrane had 0.93 m^2 of active surface area (A) and consisted of polyvinylidene difluoride (PVDF) material. The mixing within the reactor was provided by a stirrer and by constant backflow. The nitrate concentration varied between 68.5 and 75.6 mg/L in the inflow, whilst the C/N ratio was constant at 3:1. The pH level and the temperature within the reactor were controlled continuously by using an automatic control unit. The operating temperature of the 60 L reactor was within the range of 23.7-29.0 °C, with a pH value of around 8.0. Anoxic conditions within the reactor were ensured by flushing with nitrogen gas. The denitrifying culture was taken from a local wastewater treatment plant and adapted for 20 days under anoxic conditions.

Analytical procedures

The samples for analyses were taken from influent and efluent, and were collected daily. Several measurements were obtained in order to follow the denitrification efficiency: temperature, pH, the concentration of nitrate and nitrite ions, the chemical oxygen demand (COD), and the concentration of activated sludge (mixed liquor suspended solids, $MLSS$). The average values from this series of measurements were calculated at different flow rates. All the measurements were performed in accordance with ISO standards. Analyses for nitrate and nitrite concentrations were conducted spectrophotometrically [4,5] by using an Agilent 8453 UV-Visible spectrophotometer at 324 and 540 nm wavelengths, respectively. The chemical oxygen demand (COD) was determined by a volumetric method using titration with $KMnO_4$ (determination of permanganate index) [3]. $MLSS$ concentrations were measured occasionally according to standard methods for the examination of water and wastewater [6].

Calculations

The nitrate removal efficiency, R (%), and the specific denitrification rate, S (g/LNO_3/(g/L MLSS d)), were determined in order to evaluate the suitability of the membrane bioreactor for drinking water denitrification. Moreover, the MBR system's nitrate removal capacity, E_{MBR} (g/(m^2 h)), and the reactor's nitrate removal capacity, E_{R} (g/(m^3 h)), were calculated. The equations used for the calculations were taken from the literature [23].

The MBR system nitrate removal capacity (E_{MBR}) is defined as the product of the permeate flux ($J_{\mathrm{P}} = q/A$) and the difference between the average inflow and outflow nitrate concentrations:

$$E_{\mathrm{MBR}} = J_{\mathrm{P}}(\gamma(\mathrm{NO_3})_0 - \gamma(\mathrm{NO_3})_t) \quad (1)$$

Reactor nitrate removal capacity, E_{R}, (Eq. (3)) is a function of the dilution rate, D (h^{-1}), (Eq. (2)), which is defined as the quotient of the influent flow rate (q) and the bioreactor volume (V).

$$D = q / V \quad (2)$$

$$E_{\mathrm{R}} = D(\gamma(\mathrm{NO_3})_0 - \gamma(\mathrm{NO_3})_t) \quad (3)$$

A specific denitrification rate was calculated according to Eq. (4):

$$S = (\gamma(\mathrm{NO_3})_0 - \gamma(\mathrm{NO_3})_t) / (MLSS * HRT) \quad (4)$$

where $\gamma(\mathrm{NO_3})_0$ and $\gamma(\mathrm{NO_3})_t$ are the average nitrate concentrations (g/L) in the inflow and outflow during each experiment, and HRT is the hydraulic retention time (h).

The nitrate removal efficiency, R (%), was determined as:

$$R = 100(\gamma(\mathrm{NO_3})_0 - \gamma(\mathrm{NO_3})_t) / (\gamma(\mathrm{NO_3})_0 \quad (5)$$

RESULTS AND DISCUSSION

Nitrate removal efficiency and the specific denitrification rate

Eight experiments were performed, each according to the selected flow rate. The experimental results are gathered in Table 1. The results represented herein were obtained when reaching the steady-state at the selected flow rate. The average nitrate and nitrite concentrations at the inflow (marked as "in") and at the outflow (marked as "out") are shown in Figure 1. It can be seen that a relatively high level of MBR efficiency was achieved at flow rates lower than 4.8 L/h as the outflow nitrate concentrations were lower than 7 mg/L. The nitrite concentrations within the inflow were below the permitted limit value (0.5 mg/L) throughout the entire experiment. The nitrite concentrations remained below 0.5 mg/L at flow rates lower than 4.8 L/h at the outflow, whilst at higher flow rates the measurements exceeded the threshold, and the denitrification process was less effective. The microorganisms had insufficient time to complete the reaction due to the higher water-flow through the

Table 1. Experimental results depending on the inflow rates

Exp. No.	q / L h^{-1}	J_{P} / L (m^2 h)$^{-1}$	HRT / h	$MLSS$ / g L^{-1}	COD / mg L^{-1}
1	0.6	0.6452	100.0	0.7731	0.51
2	1.2	1.2903	50.0	0.7693	0.77
3	1.8	1.9355	33.3	0.7983	0.79
4	2.4	2.5806	25.0	0.8312	0.96
5	3	3.2258	20.0	0.6031	1.07
6	4.8	5.1613	12.5	0.8090	1.12
7	7.2	7.7419	8.3	0.9327	3.10
8	10.2	10.9677	5.9	0.7668	7.02

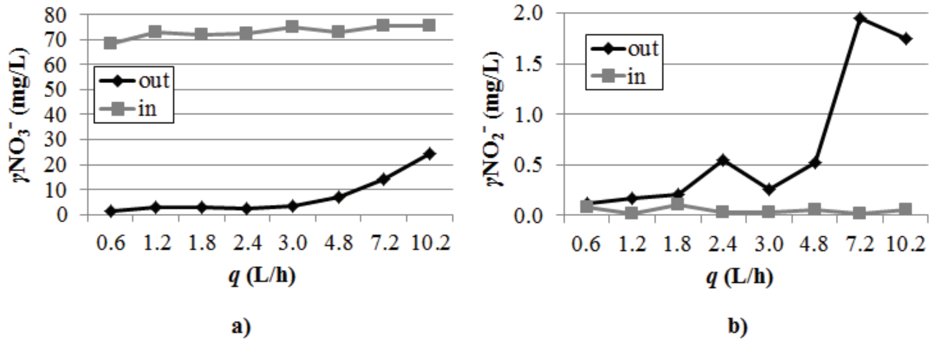

Figure 1. Average nitrate (a) and nitrite (b) concentrations measured at the inflow and at the outflow, depending on the flow rate (for eight different experiments).

reactor and therefore lower hydraulic retention time. Consequently, the concentrations of nitrates and nitrites increased at the outflow.

The highest removal efficiency, *R* (%), was achieved at the lowest inflow rates (Figure 2). When the flow rate varied within the range from 0.6 to 4.8 L/h, the removal efficiency was above 90%, whilst at the lowest inflow rate used the efficiency was even 97.67%. The hydraulic retention time (*HRT*) decreased with any further increase of the inflow and the removal efficiency consequently decreased. This is in accordance with the findings of previous studies, which confirmed that higher HRT values usually result in better removal performance and lower values indicate higher organic loading rates [22]. On the other hand, short HRT requires a high trans-membrane flux and could influence the quality of the treated effluent [15]. The experiments performed during this research indicated that denitrification efficieny (nitrate removal) was also affected by the oxygen and pH variations, as well as the inhibitive effect of accumulated nitirite at higher flow rates that could not be neglected even if the amounts were very low. Nevertheless, sufficient results were obtained at the highest inflow of 10.2 L/h (dilution rate 0.17 h^{-1}, *HRT* 5.9 h), namely 68.08% of the nitrate was removed. Although the nitrate was incompletely removed, the efficiencies obtained from this study can be compared with the results of other studies using extractive denitrifying MBR [21]. The nitrate removal efficiency was above 99% with an initial nitrate concentration of 200 mgNO_3/L using methanol as substrate, and denitrification rates were obtained of up to 1.1 g/(m^2 d). In another research performed by two-stage anoxic/oxic biofilm MBR with ethanol (C/N ratio = 1.4-2.5) and commercially available Biocontact-N biocarriers (to enable immobilisation), the nitrate conversions were also very high. In addition, no nitrite formation was observed during the process, whereas the influent nitrate concentration was equal to 150 mg/L (*HRT* 2.5 h) [20]. Similarly, a varying degree of nitrate reduction, from 96% to complete removal, was found in the denitrification of water with 60 mg/L NO_3 in the inflow [7].

Significant impact on the specific denitrification rate was observed by the variations of flow rates (dilution rates). The maximum specific denitrification rate was acquired at the highest inflow rate of 10.2 L/h (Figure 2) and reached the value of 0.2738 g/L NO_3/(g/L MLSS d). According to the calculated values and Eq. (4), the specific denitrification rate increases by decreasing the *HRT*. The higher the dilution rate, the higher the denitrification rate.

Similar results were achieved during the research performed on ground water, as performed by Buttiglieri [19], using ethanol as the carbon source (C/N ratio = 2.2, *HRT* 19-37 h), whereas the inflow nitrate concentration of 30 mg/L allowed maximum nitrate removal rates of between 0.36 and 0.48 gNO_3/(gTSS d). It was reported that the bench scale MBR was able to offer nitrate removals of up to 98.5%, which is close to the values obtained herein, whilst the specific denitrification rates achieved were lower, up to 0.02 d^{-1} [23]. There are several ways of explaining the differences in the above-mentioned results. The type and amount of external carbon source might significantly affect the microbial growth and its activity, which could be further reflected in lower nitrate removal and denitrification rates [10].

The concentrations of mixed-liquor suspended solids (MLSS) within the membrane bioreactor varied at between 0.6031 and 0.9327 g/L. This value was lower by half when compared to study of nitrate removal from synthetic groundwater prepared by lake water and performed within same type of MBR, where anoxic sludge concentrations were determined of between 1.6 and 2 g/L [19]. Although the MLSS concentration was low, the problem of fouling was observed during the final phase of the MBR operation

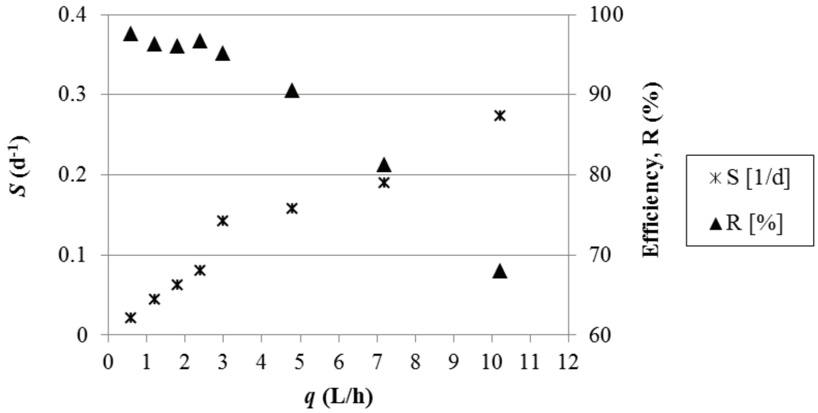

Figure 2. Specific denitrification rate and removal efficiency depending on the flow rate.

and the transmembrane pressure increased. Since the denitrifying culture accumulates on the membrane surface where it forms filter cake, the membrane separation efficiency decays over time. One of the reasons that has been reported to cause membrane fouling in MBRs is decreasing *HRT* [22]. The membrane fouling could afterwards lead to increased energy consumption and operating costs [13]. Therefore, appropriate measures need to be ensured to prevent membrane fouling. It could be additionaly affected by sludge retention time and the characteristics of activated sludge [22]. Based on these facts, it can be concluded that the problem of fouling during our experiments could have been caused by decreased *HRT*, since the MLSS concentration was low and did not increase significantly.

The biological denitrification of drinking water performed by heterotrophic micro-organisms requires an aditional carbon source, although residual carbon sources in effluent can cause many problems during drinking water treatment [9]: fast growth of bacteria, formation of by-products within the treated water, and high *COD* values in the efluent [11]. In this experiment the values of *COD* measurements in the effluent (Table 1) were below 1 mg O_2/L at lower inflow rates, while the maximum value of 7.02 mg O_2/L, which exceeds the threshold value of 5 mg O_2/L was achieved at the highest inflow rate. However, the concentrations at the inflow were around 95 mg O_2/L due to the added carbon source, *i.e.*, sucrose. Although sucrose in studies on denitrification has been rarely used as a carbon source, the presented study proved it to have quite good potential for nitrate removal efficiency.

Reactor removal capacity

In order to facilitate the evaluation of the experimental results and to easily compare the tested MBR system, it was necessary to calculate the reactor's removal and the MBR system's removal capacities. The reactor removal capacity (E_R) of the pilot--scale MBR was calculated in accordance with Eq. (3). A maximum reactor removal capacity of 8.75 g/(m^3 h) was achieved at the highest dilution rate (Figure 3). This could be explained by the fact that these two parameters are linearly correlated. Therefore, the higher the dilution rate, the higher the reactor removal capacity. The lowest capacity achieved by MBR was below 1 g/($m^{3}\cdot$h). The study on experimental results demonstrated that the capacity not only depends on nitrate removal and the volume of the reactor, but also on the process conditions, such as flow rate, HRT, etc. However, according to Figure 3, it appears that the best performances of the reactor system can be obtained if the flow rate is higher than 4.8 L/h. But it must be considered that at higher flow rates denitrification was incomplete due to the lower *HRT*s, and therefore the nitrate removal efficiencies were much lower. In addition, the concentration of the effluent *COD* at the highest flow rate was above the value limit. Therefore, operation at a flow higher than 4.8 L/h is unreasonable. The optimal reactor removal capacity (E_R) that could be accepted regarding the above mentioned facts for these experiments was achieved at 5.29 g/(m^3 h). The reactor removal capacities obtained herein are up to eight times higher due to the more than ten times higher inflow rate, compared with the literature [23]. Moreover, in this research the active surface area of the membrane and the working volume of the reactor, were considerably higher. Otherwise, the removal capacity during nitrate removal from the ground water using methanol as the carbon source and at the inflow a nitrate concentration of 60 mg/L, was higher than that calculated in the presented study (29.2-70.8 gNO_3/(m^3 h)) [7].

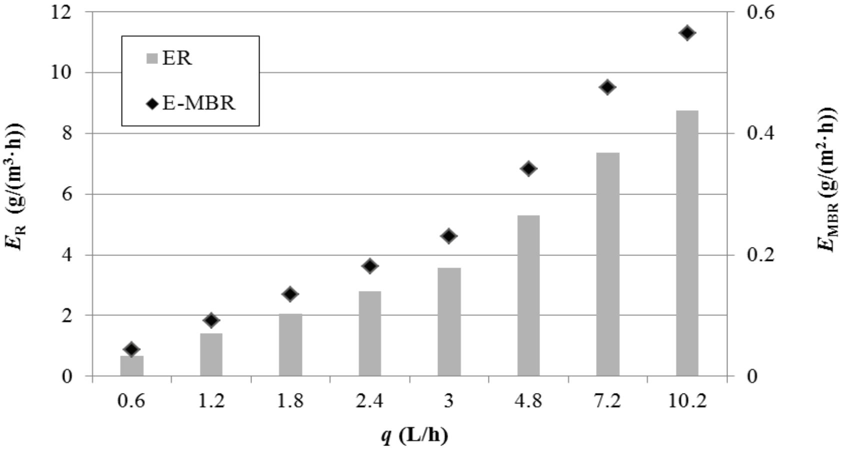

Figure 3. The reactor removal capacity and MBR system removal capacity depending on the flow rates.

Finally, the MBR system nitrate removal capacity (E_{MBR}) was calculated according to Eq. (1). The MBR removal capacity is highly influenced by the type of filtration membrane and consequently by the active surface area of the membrane. The lowest value 0.04 g/(m^2·h) was obtained at an inflow rate of 0.6 L/h, whilst the maximum value 0.56 g/(m^2·h) was calculated at the highest inflow rate (dilution rate).

The capacities for both the reactor and MBR system increased linearly with the flow rate (see Figure 3). However, by increasing the flow rate the capacity can increases but this could have a negative influence on the nitrate removal efficiency, as shown above. Similarly as for the reactor removal capacity, it could be concluded that optimal MBR system capacitiy was achieved at a flow rate of 4.8 L/h at 0.34 g/(m^2 h). It was found also that the ratio of MLSS and the initial nitrate concentration at the inflow, had had insignificant impact on the capacity and therefore with the lower mass ratio higher specific denitrification rates could be achieved [23]. The MBR system removal capacities calculated during this experiment were close to the results obtained in the above-mentioned paper, just slightly lower efficinces were determined due to the lower permeate flux and the differences in the specifications of the membrane module.

However, efficient denitrification in MBR using sucrose as the carbon source and at C/N ratio at 3:1 was visible at flow rates lower than 4.8 L/h (dilution rate 0.08 h^{-1}). Nitrate removal efficiency exceeding 90% could be expected under these operational conditions. The drawback is that the removal capacities at low flow rates were low. In contrast, removal capacities were very high at above 4.8 L/h, but incomplete denitrification results in the production of nitrite and increase of *COD*. Therefore, the presented MBR system is beneficial for application within small water systems that operate at low flow rates.

CONCLUSIONS

The suitability of the membrane bioreactor for drinking water denitrification was investigated in this research. It was found that:

- Denitrification of drinking water in the pilot-scale MBR by using sucrose as the carbon source successfully removed nitrates from the drinking water.
- High nitrate removal efficiencies (more than 90%) were obtained at flow rates below 4.8 L/h.
- The denitrification efficiencies were highly dependent on the operational conditions, especially the flow rate, and therefore the hydraulic retention time.
- At lower HRTs, denitrification was incomplete and an accomulation of nitrite was observed.

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ALEKSANDRA PETROVIČ
MARJANA SIMONIČ

Faculty of Chemistry and Chemical Engineering, University of Maribor, Maribor, Slovenia

NAUČNI RAD

EFIKASNOST MEMBRANSKOG BIOREAKTORA ZA DENITRIFIKACIJU VODE ZA PIĆE

U radu je ispitan kapacitet membranskog bioreactora (MBR) za uklanjanje nitrata iz pijaće vode. Testirane su mogućnosti poluindustrijskog postrojenja MBR u zavisnosti od operativnih parametara koristeći saharozu kao izvor ugljenika. Pijaća voda iz izvora je dovedena u reaktor da bi se analizirao uticaj protoka na efikasnost denitrifikacije i uklanjanja nitrata iz pijaće vode. Sadržaj nitrata je bio oko70 mg/l, a odnos C/N je bio 3:1. Efikasnost uklanjanja nitrata je oko 90% pri protoku manjem od 4,8 l/h. Specifična brzina denitrifikacije je bila u opsegu od 0,02 do 0,16 g/l NO_3*/(g/L MLSS d). Efikasnost uklanjanja nitrata je značajno zavisila od protoka i hidrauličkog vremena zadržavanja. Pri maksimalnom protoku od 10,2 L/h moguće je uklanjanje do 68% nitrata, dok je ostvarena najveća specifična brzina denitrifikacije od 0,2738 g/L* NO_3*/(g/L) MLSS d). Izračunato je da je maksimalni kapacitet uklanjanja nitrata 8,75g* NO_3/m^3 *h.*

Ključne reči: kapacitet, denitrifikacija, pijaća voda, efikasnost, membranski bioreaktor, saharoza.