

# The influence of electric current density on specific denitrification rate of and nitrogen removal rate in electrochemical and electrobiological rotating contactor

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**Keywords:** electric current density, rotating electrochemical disk contactor, rotating electrobiological disk contactor, specific denitrification rate, nitrogen removal rate.

**Abstract:** This study aimed to determine the influence of the electric current density on the rate of nitrogen compounds removal ( $r_N$ ) and the specific rate of denitrification ( $r_D$ ) in a rotating electrochemical disk contactor (REDC) and a rotating electro-biological disk contactor (REBDC). In REBDC and REDC, the cathode consisted of disks with immobilized biomass and disk, from which biofilm was periodically removed, respectively. An aluminum anode was mounted in contactor chambers. The study was conducted using synthetic wastewater with characteristics similar to wastewater from soilless cultivation of tomatoes. The first stage of the study determined  $r_N$  and  $r_D$  in the REDC. The second stage determined  $r_N$  and  $r_D$  in the REBDC. Four hydraulic retention times (HRT) were tested: 4 h, 8 h, 12 h, and 24 h, with electric current densities of 0.63 A/m<sup>2</sup>, 1.25 A/m<sup>2</sup>, 2.50 A/m<sup>2</sup>, 5.00 A/m<sup>2</sup>, and 10.00 A/m<sup>2</sup>.

In REDC, a linear dependency was observed between  $r_N$  and current density in the examined HRTs, whereas in REBDC, a logarithmic dependency was confirmed between  $r_N$  and current density. In both contactors, an exponential dependency was observed between  $r_D$  and current density. The specific rate of denitrification decreased when the current density and HRT were increased. The study showed that, in both contactors, the rate of total nitrogen removal increased when the current density was increased and the HRT was decreased.

## Introduction

Electrochemical and bio-electrochemical (electro-biological) methods are promising solutions for the soilless plant cultivation in greenhouses, generating vast loads of phosphorus compounds and oxidized forms of nitrogen (Rodziewicz et al. 2019). They ensure a high degree of contaminants neutralization and represent the so-called friendly technology as they generate low amounts of sludge and by-products. Also, their investments and exploitation costs are low compared to these of the physicochemical processes, like ultrafiltration or ionic exchange. Simple operation as well as technological and technical reliability are important during the implementation process in agricultural facilities that have no access to technologically qualified staff.

Electrooxidation is an electrochemical process in which the electrical field is applied between particular anodes and cathodes to enable the treatment of wastewater flowing between them. Electrochemical processes can be performed in simple-construct reactors. Unlike the bio-process, electrooxidation does not require the adaptation period nor additional chemical compounds because of being selective to nitrate reduction

(Kabdaşlı et al. 2012). The mechanism of electrolytic reduction of nitrates is complex. Nitrate ions are reduced to nitrite, and ultimately to gaseous nitrogen on cathode's surface. The products of water electrolysis include gaseous hydrogen and oxygen. Nitrates are very well soluble in water and generate a few types of by-products (Li et al. 2009). Nitrites and ammonia are the main by-products of electrochemical nitrate reduction (Li et al. 2009, Huang et al. 2013)

The most frequent problem encountered during electrochemical nitrate reduction includes the generation of such by-products as nitrites, ammonia, and hydroxylamines, which are considerably more toxic than nitrates being removed (Dash and Chaudhari 2005, Polatides et al. 2005, Katsounaros and Kyriacou 2008). Despite these limitations, this is still a highly effective method for nitrate removal, as evidenced by Govindan et al. (2015). They demonstrated 92% effectiveness of nitrate removal for an aluminum anode and an iron cathode, as well as 80% effectiveness for the system with an iron anode and cathode, at electric current density of 25 mA/cm<sup>2</sup>, electrolysis duration of 180 minutes, and initial nitrate concentration of 100 ppm. However, regardless of cathode material, the electrochemical reduction is always

coupled with electrocoagulation, leading to ammonia synthesis. Dissolving anodes (Fe and Al) also contribute to nitrates removal in the electrocoagulation process (Koparal and Öütveren 2002, Lacasa et al. 2011, Yehya et al. 2015, Elazzouzi et al. 2019).

Denitrification performed in a bio-electrochemical reactor is based on the activity of autohydrogenotrophs in the biofilm and requires employing an appropriate method for providing these microorganisms with hydrogen. The blowing of gaseous hydrogen into the reactor was the first method used to this end. However, it bears some limitations considering mainly a low hydrogen solubility in an aqueous solution (1.6 mg/l at 20°C) (Lee and Rittmann 2002). This hydrogen is little available for microorganisms, which consequently results in lower nitrate removal rates compared to heterotrophic denitrification (Prosnansky et al. 2002, Vasiliadou et al. 2006). Other drawbacks of this method include, e.g., high costs of feeding the system with gaseous hydrogen and the production of explosive H<sub>2</sub> mixtures with air. A solution to these problems has been sought in the proposed *in situ* techniques of hydrogen production (Feleke and Sakakibara 2002, Lee and Rittmann 2002), including, e.g., water electrolysis. The rate of this common hydrogen production method can be precisely controlled by the inflow of electric current (Prosnansky et al. 2002). A new type of reactor, called a bio-electrochemical reactor – BER, offers the coupling of electric energy with bio-denitrification. The surface of its cathode ensures the site for denitrifying microorganisms immobilization, owing to which they can be in direct contact with *in situ* produced hydrogen (Watanabe et al. 2001).

The anode used in BER can be made of, e.g., amorphous carbon (Sakakibara et al. 1997), platinum-covered titanium (Prosnansky and Sakakibara 2002), or modified  $\beta$ -PbO<sub>2</sub> (Zhou et al. 2007), whereas cathodes can be made of carbon (Feleke et al. 1998, Islam and Suidan 1998), graphite felt (Park et al. 2005), and steel (Sakakibara et al. 1997).

Two parameters may characterize the effectiveness of nitrates removal in electrochemical and electro-biological reactors. The first one is the rate of nitrogen compounds removal ( $r_N$ ), which indicates the amount of nitrogen load removed per cathode surface area (Kłodowska et al. 2015). The

second one is the specific rate of nitrogen compounds removal, also referred to as the specific rate of denitrification ( $r_D$ ), which indicates nitrogen load removed using 1 kWh. It needs to be remembered that in the electrochemical reactor (with disks not covered with a biofilm), the removal of nitrogen compounds proceeds via electrocoagulation and electrochemical reduction of nitrates. In contrast, in the electro-biological reactor (with disks covered with a biofilm) it is due not only to electrocoagulation and the electrochemical reduction of nitrates but also to hydrogenotrophic and heterotrophic denitrification and biofilm biomass growth.

This study aimed to determine the influence of electric current density on the rate of nitrogen compounds removal ( $r_N$ ) and the specific rate of denitrification ( $r_D$ ) in an electrochemical rotating contractor and an electro-biological rotating contractor.

## Materials and methods

The experiments were divided into two parts. The first part aimed to determine the rate of nitrogen compounds removal and the specific rate of denitrification in the RECDC (rotating electrochemical disk contactor) with disks not covered with a biofilm, as a result of electrochemical reduction of nitrates. Over the experimental period, the disks were regularly cleansed to remove developing microorganisms of the biofilm. Nitrogen compounds were removed during electrocoagulation and electrochemical reduction of nitrates. The second part aimed to determine the rate of nitrogen compounds removal and the specific rate of denitrification in the REBDC (rotating electro-biological disk contactor) with disks covered with a biofilm. In the REBDC, nitrogen compounds were removed during hydrogenotrophic and heterotrophic denitrification, electrocoagulation and electrochemical reduction of nitrates, and biomass growth.

The reactor used in the study was a rotating biological disk contactor in which stainless steel disks served as a cathode, and an aluminum electrode mounted in the flow-tank of the contactor served as an anode (Fig. 1).

Both electrodes were connected to a laboratory power supply (HANTEK PPS2116A, China), which served as

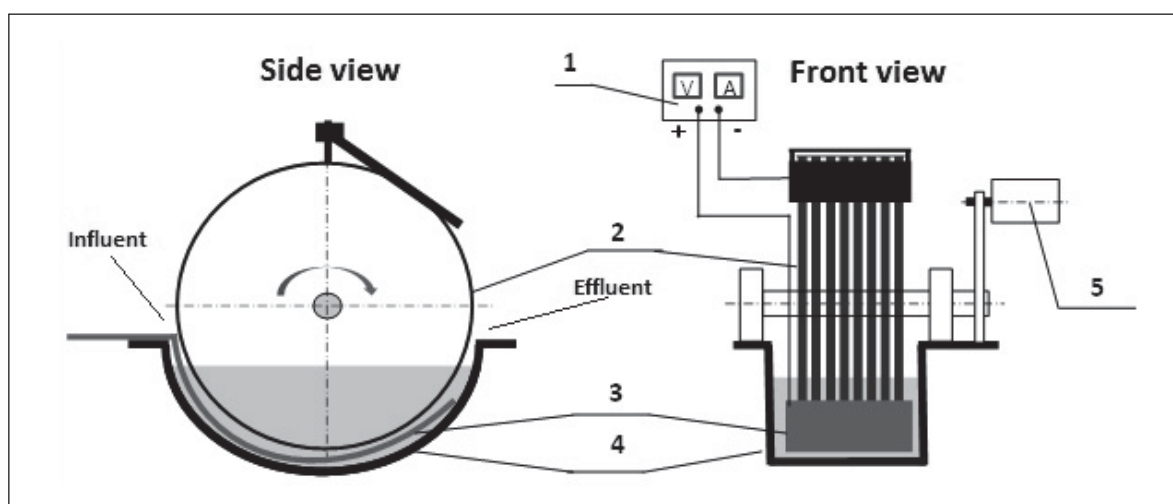


Fig. 1. Scheme of the single-stage rotating disk contactor: 1 – laboratory power supply, 2 – discs (cathode), 3 – anode, 4 – tank, 5 – electric engine (Rodziewicz et al. 2019)

a source of direct electric current. The experiment was carried out in a laboratory scale on four single-stage rotating disk contactors (8 disks with a diameter of 0.22 m; flow chamber volume of 2.0 L).

Experiments were conducted at a temperature of about  $20.0 \pm 1^\circ\text{C}$ ; four hydraulic retention times: 4 h, 8 h, 12 h, and 24 h; and the following intensities of electric current: 0.35 A, 0.70 A, 1.40 A, 2.80 A and 5.60 A (electric current density of:  $0.63 \text{ A/m}^2$ ,  $1.25 \text{ A/m}^2$ ,  $2.50 \text{ A/m}^2$ ,  $5.00 \text{ A/m}^2$ , and  $10.00 \text{ A/m}^2$ , respectively). The density values of electric current were adjusted based on the literature data (Zhao et al. 2011). Synthetic wastewater with the composition similar to that of wastewater from the soilless cultivation of tomatoes, adopted after Saxena and Bassi (2013) and Mielcarek et al. (2019) was used. Sodium acetate was added to wastewater as a source of carbon in part II of the study to provide appropriate conditions for biofilm development. This allowed for obtaining the C: N ratio of 0.5, which promotes the development of heterotrophic biofilm organisms and, at the same time, does not impair autotrophic denitrification (Rodziewicz et al., 2015) the introduction of external carbon sources becomes necessary. Wastewater characteristic is presented in Table 1.

Wastewater was fed to reactors with Minipuls 3 peristaltic pumps (Gilson, USA) and analyzed using the following indicators:

- total nitrogen – using a Total Organic Carbon Analyzer TOC-L CPH/CPN with TNM-L device (Shimadzu Corporation, Japan), with a method of oxidative combustion – chemiluminescence;
- ammonia nitrogen, nitrate nitrogen, and nitrite nitrogen – using a spectrophotometer, with a colorimetric method;
- pH value ( $\pm 0.01 \text{ pH}$ ), temperature ( $\pm 1^\circ\text{C}$ ) – using a CP-105 pH meter (Elmetron, Poland); redox potential ( $\pm 1 \text{ mV}$ ) – with a pH 211 meter (Hanna Instruments, Spain); electrolytic conductivity ( $\pm 0.01 \text{ mS/cm}$ ) – using an HQ 440d multi-meter (Hach Company, USA);

In the second part of the study, the analytic control of processes was begun after reactor adaptation. In this part of the study, the activated sludge from denitrification tanks of the Municipal Wastewater Treatment Tanks “Łyna” in Olsztyn was used as the inoculum.

Samples were collected for analyses in 24-h intervals. In both parts of the study, the results of physicochemical analyses of 20 samples were used to calculate the average value.

The rate of nitrogen compounds removal ( $r_N$ ) was computed from the following formula:

$$r_N = \frac{Q(C_{No} - C_{Ne}) \cdot 24}{A} [\text{mg N}/(\text{cm}^2 \cdot \text{d})] \quad (1)$$

where:

$C_{No}$  – concentration of total nitrogen in the inflowing wastewater [mg N/L],

$C_{Ne}$  – concentration of total nitrogen in the effluent [mg N/L],

$Q$  – flow rate [L/h],

$A$  – total surface area of the cathode [ $\text{cm}^2$ ],

24 – a conversion factor of hours into days;

In turn, the specific rate of nitrogen compounds removal (specific rate of denitrification), which denotes the load of total nitrogen removed using 1 kWh, was computed from the following formula:

$$r_D = \frac{Q(C_{No} - C_{Ne})}{UI} [\text{mg N}/(\text{kWh})] \quad (2)$$

where:

$C_{No}$  – concentration of total nitrogen in the inflowing wastewater [mg N/L],

$C_{Ne}$  – concentration of total nitrogen in the effluent [mg N/L],

$Q$  – flow rate [L/h],

$I$  – intensity of electric current used in the experiment [mA],

$U$  – voltage applied [V].

## Results and discussion

### *The rate of nitrogen compounds removal ( $r_N$ ) in a rotating electrochemical disk contactor*

A linear dependency was observed in the electrochemical process of wastewater treatment between total nitrogen removal rate ( $r_N$ ) and electric current density in the examined range of HRTs (Fig. 2), which was confirmed by a high value of the determination coefficient ( $R^2$ ). The linear relationship applies only to the current density range tested in the study ( $0.63\text{--}10.0 \text{ A/m}^2$ ).

**Table 1.** Synthetic wastewater concentration

Parameters	Mean	Minimum	Maximum	Standard deviation
COD [ $\text{mg O}_2/\text{L}$ ]*	260	218	318	40
COD [ $\text{mg O}_2/\text{L}$ **]	45	37	57	6
Total nitrogen [mg N/L]	470	446	490	15
Nitrate [mg N/L]	453	429	469	15
Ammonia nitrogen [mg N/L]	18	17	21	2
Nitrite [mg N/L]	0.015	0.010	0.020	0.002
Total phosphorus [mg P/L]	74	49	85	10
pH	6.98	6.85	7.20	–
Electrolytic conductivity [mS/cm]	6.0	5.7	6.3	0.2
Redox potential [mV]	265	249	278	26

\* wastewater with sodium acetate

\*\* wastewater without sodium acetate

At HRT of 4 h, the rate of nitrates removal increased from 0.1 mg N/(cm<sup>2</sup>·d) to 0.2 mg N/(cm<sup>2</sup>·d) at electric current densities of 0.63 A/m<sup>2</sup> and 10.00 A/m<sup>2</sup>, respectively. Under the flow of electric current with densities of 1.25 A/m<sup>2</sup>, 2.50 A/m<sup>2</sup>, and 5.00 A/m<sup>2</sup>, the respective rates reached 0.11 mg N/(cm<sup>2</sup>·d), 0.12 mg N/(cm<sup>2</sup>·d), and 0.15 mg N/(cm<sup>2</sup>·d).

At HRT of 8 h and electric current density of 0.63 A/m<sup>2</sup>, the rate of nitrogen compounds removal reached 0.07 mg N/(cm<sup>2</sup>·d), whereas at current densities of 1.25 A/m<sup>2</sup>, 2.50 A/m<sup>2</sup>, and 5.00 A/m<sup>2</sup>, the respective removal rates were 0.07 mg N/(cm<sup>2</sup>·d), 0.08 mg N/(cm<sup>2</sup>·d), and 0.1 mg N/(cm<sup>2</sup>·d). At the highest density of electric current,  $r_N$  reached 0.14 mg N/(cm<sup>2</sup>·d). Extending HRT to 12 h led to a successive increase in the nitrogen compounds removal rate. At this HRT and electric current densities increasing in the analyzed range, the rate of nitrogen load removal increased as well. At electric current densities of: 0.63 A/m<sup>2</sup>, 1.25 A/m<sup>2</sup>, 2.50 A/m<sup>2</sup>, 5.00 A/m<sup>2</sup>, and 10.00 A/m<sup>2</sup>, the respective nitrogen removal rates were as follows: 0.06 mg N/(cm<sup>2</sup>·d), 0.07 mg N/(cm<sup>2</sup>·d), 0.07 mg N/(cm<sup>2</sup>·d), 0.09 mg N/(cm<sup>2</sup>·d), and 0.12 mg N/(cm<sup>2</sup>·d). At the longest, 24-hour hydraulic retention time and electric current densities of 10.00 A/m<sup>2</sup> and 5.00 A/m<sup>2</sup>, the rates of nitrogen load removal reached 0.09 mg N/(cm<sup>2</sup>·d) and 0.06 mg N/(cm<sup>2</sup>·d), respectively. Whereas at this HRT and the other current densities tested (0.63 A/m<sup>2</sup>, 1.25 A/m<sup>2</sup>, 2.50 A/m<sup>2</sup>), nitrogen was removed with the rate of 0.04 mg N/(cm<sup>2</sup>·d).

#### The rate of nitrogen compounds removal in a rotating electro-biological disk contactor

In HRT range from 4 h to 24 h, a logarithmic dependency was demonstrated between the rate of total nitrogen removal ( $r_N$ ) and electric current density (Fig. 3), as confirmed by a high value of the determination coefficient ( $R^2$ ). This logarithmic correlation is due to many different processes that take place in the electro-biological reactor. The electric current flow results in nitrate electrocoagulation, electrochemical nitrate reduction, and hydrogenotrophic (autotrophic) denitrification. But, heterotrophic denitrification proceeds in

parallel. Each of these three processes occurs at a different rate. The equations presented in the graphs again describe the course of the curve only for current densities ranging from 0.63 to 10.0 A/m<sup>2</sup>.

At the shortest HRT – 4 h, nitrogen removal rate increased to 0.13 mg N/(cm<sup>2</sup>·d) at  $J = 0.63$  A/m<sup>2</sup>, and to 0.31 mg N/(cm<sup>2</sup>·d) at  $J = 10.00$  A/m<sup>2</sup>. Under the flow of electric current with densities of 1.25, 2.50, and 5.00 A/m<sup>2</sup>, the removal rate increased to 0.15 mg N/(cm<sup>2</sup>·d), 0.21 mg N/(cm<sup>2</sup>·d), and 0.24 mg N/(cm<sup>2</sup>·d), respectively. Extending HRT to 8 h caused a decrease in nitrogen removal rate. At the lowest current density ( $J = 0.63$  A/m<sup>2</sup>), the removal rate reached 0.08 mg N/(cm<sup>2</sup>·d). A successive increase in electric current density resulted in a nitrogen removal rate increase. At densities of 1.25 A/m<sup>2</sup>, 2.50 A/m<sup>2</sup>, and 5.00 A/m<sup>2</sup>, the respective rates accounted for 0.1 mg N/(cm<sup>2</sup>·d), 0.13 mg N/(cm<sup>2</sup>·d), and 0.15 mg N/(cm<sup>2</sup>·d). The highest removal rate, i.e., 0.19 mg N/(cm<sup>2</sup>·d), was achieved at  $J = 10.00$  A/m<sup>2</sup>.

When the hydraulic retention time was 12 h and the current density was  $J = 0.63$  A/m<sup>2</sup>, nitrogen compounds were removed with the rate of 0.06 mg N/(cm<sup>2</sup>·d). The removal rate was observed to increase along with increasing density of the electric current and to reach 0.08 mg N/(cm<sup>2</sup>·d), 0.11 mg N/(cm<sup>2</sup>·d), and 0.13 mg N/(cm<sup>2</sup>·d) at  $J = 1.25$  A/m<sup>2</sup>,  $J = 2.50$  A/m<sup>2</sup>, and  $J = 5.00$  A/m<sup>2</sup>. The highest removal rate, reaching 0.17 mg N/(cm<sup>2</sup>·d), was achieved at  $J = 10.00$  A/m<sup>2</sup>. Nitrogen compounds were removed with the lowest rate ( $r_N = 0.04$  mg N/(cm<sup>2</sup>·d)), at HRT = 24 h and  $J = 0.63$  A/m<sup>2</sup>. At the higher electric current densities, i.e., 1.25 A/m<sup>2</sup>, 2.50 A/m<sup>2</sup>, 5.00 A/m<sup>2</sup>, and 10.00 A/m<sup>2</sup>, nitrogen load removal rates reached: 0.05 mg N/(cm<sup>2</sup>·d), 0.06 mg N/(cm<sup>2</sup>·d), 0.08 mg N/(cm<sup>2</sup>·d), and 0.12 mg N/(cm<sup>2</sup>·d), respectively.

A comparison of nitrogen compounds removal rates in RECD and REBDC reactors indicates that at HRT = 4 h, the  $r_N$  was higher in REBDC than in RECD, regardless of electric current density. An increase in current density caused a growing difference in the nitrogen removal rate between

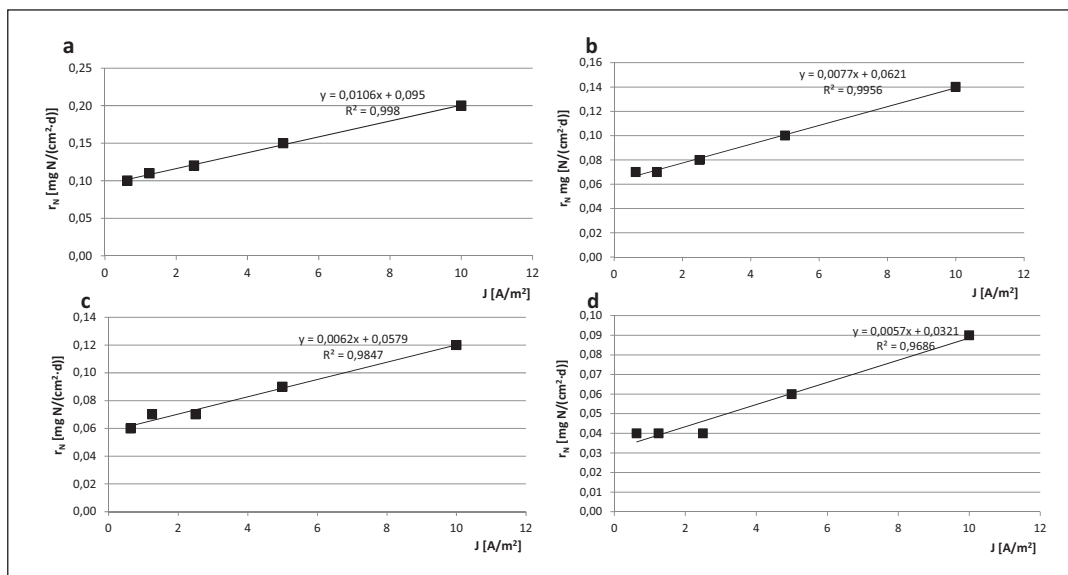


Fig. 2. The rate of nitrogen compounds removal in rotating electrochemical disk contactor, depending on the electric current density: a – HRT = 4 h, b – HRT = 8 h, c – HRT = 12 h, d – HRT = 24 h

the reactors. At electric current density of  $0.63 \text{ A/m}^2$ , the  $r_N$  determined in REBDC was higher by 30% than in RECDC ( $0.13 \text{ mg N}/(\text{cm}^2\cdot\text{d})$  and  $0.10 \text{ mg N}/(\text{cm}^2\cdot\text{d})$ , respectively). At the highest current density of  $10.0 \text{ A/m}^2$ , the  $r_N$  was higher by as much as 55% in REBDC than in RECDC ( $0.31 \text{ mg N}/(\text{cm}^2\cdot\text{d})$  and  $0.20 \text{ mg N}/(\text{cm}^2\cdot\text{d})$ , respectively). HRT extension diminished differences in nitrogen removal rates between the electrochemical and the electro-biological reactors. At  $J = 0.63 \text{ A/m}^2$  and  $\text{HRT} = 12 \text{ h}$  and  $\text{HRT} = 24 \text{ h}$ , the removal rates were the same. In turn, at  $J = 10.0 \text{ A/m}^2$ , the  $r_N$  was higher by 33% in REBDC than in RECDC ( $0.12 \text{ mg N}/(\text{cm}^2\cdot\text{d})$  and  $0.09 \text{ mg N}/(\text{cm}^2\cdot\text{d})$ , respectively). Islam and Suidan (1998) achieved a comparable rate of nitrates removal at  $0.8 \text{ g N/d}$ , per  $1 \text{ m}^2$  of electrode surface  $0.08 \text{ mg N}/(\text{cm}^2\cdot\text{d})$ ). They tested hydraulic retention times from 10 to 13 h in an electrochemical reactor with the specific surface of the electrode reaching  $42 \text{ m}^2/\text{m}^3$ . In the bio-electrochemical reactor with an electrode having the surface area of  $321 \text{ cm}^2$ , Wang and Qu (2003) reached lower denitrification rates ( $0.034\text{--}0.43 \text{ mg N-NO}_3/(\text{cm}^2\cdot\text{d})$ ) than Park et al. (2006), who used an electrode having the surface area of  $105 \text{ cm}^2$ . This indicates that electrode's surface and specific surface are important parameters influencing denitrification effectiveness, and that a longer hydraulic retention time (HRT from 10 h to few days) is needed for complete denitrification (Sakakibara et al. 1995, Feleke et al. 1998, Islam and Suidan 1998).

The study showed that in the electro-biological rotating contactors, the rate of total nitrogen removal increased when the current density was increased and the HRT was decreased. In the electro-biological reactor, extending HRT increased the efficiency of the denitrification process, which increases  $\text{OH}^-$  concentration, pH value, and, consequently, nitrite concentration, which may lead to denitrification process inhibition by the accumulation of nitrites. According to the literature (Shin et al. 2005; Zhou et al. 2007), the optimal pH for the denitrification process is  $7.5\text{--}7.6$ , and denitrification is inhibited above these values due to the accumulation of nitrites. In the study by Rodziewicz et al. (2019), wastewater pH in the

reactor exceeded 8.2 for all HRT values and current densities tested. The increase in the pH value in the reactor is due to the denitrification process itself. However, alkalinity may also increase due to the use of bio-electrochemical reactors. The preparation of 1 mole of  $\text{H}_2$  gives two moles of  $\text{OH}^-$ . In addition, during the denitrification process, the removal of one mole of nitrates involves the formation of one mole of  $\text{OH}^-$ , which increases the pH value, and thus causes nitrite accumulation and reduces the rate of nitrate removal (Ghafari et al. 2009; Mousavi et al. 2012).

### Specific rate of denitrification ( $r_D$ ) in a rotating electrochemical disk contactor

In this study, we also computed the specific rate of denitrification, which indicates the amount of total nitrogen removed at an energy consumption of 1 kWh (Fig. 4). At hydraulic retention times of 4 h and 24 h, an exponential dependency was demonstrated between the specific rate of denitrification ( $r_D$ ) and electric current density, as evidenced by high values of the determination coefficient ( $R^2$ ).

The specific rate of total nitrogen removal decreased along with an increasing density of electric current and with extended hydraulic retention time. It was  $6.4 \text{ mg N/kWh}$  at the shortest  $\text{HRT} = 4 \text{ h}$  and the lowest  $J = 0.63 \text{ A/m}^2$ . Under the flow of electric current with a density of  $10.00 \text{ A/m}^2$ , the specific rate of denitrification reached  $0.85 \text{ mg N/kWh}$ . At  $\text{HRT} = 8 \text{ h}$ , the  $r_D$  was  $4.46 \text{ mg N/kWh}$  and  $0.58 \text{ mg N/kWh}$  at the lowest and highest current densities, respectively. Extending hydraulic retention time to 12 h decreased the load of total nitrogen removed. In the analyzed range of electric current densities, i.e.,  $0.63\text{--}10.00 \text{ A/m}^2$ , specific rates of denitrification ranged from  $4.34$  to  $0.52 \text{ mg N/kWh}$ . At the longest hydraulic retention time, i.e.,  $\text{HRT} = 24 \text{ h}$ , the  $r_D$  was observed to decrease successively. It reached  $2.47 \text{ mg N/kWh}$  and  $0.38 \text{ mg N/kWh}$  at the lowest and highest current densities, respectively. At electric current densities of  $1.25 \text{ A/m}^2$ ,  $2.50 \text{ A/m}^2$ , and  $5.00 \text{ A/m}^2$ , the  $r_D$  values achieved were  $-1.35 \text{ mg N/kWh}$ ,  $0.72 \text{ mg N/kWh}$ , and  $0.47 \text{ mg N/kWh}$ , respectively.

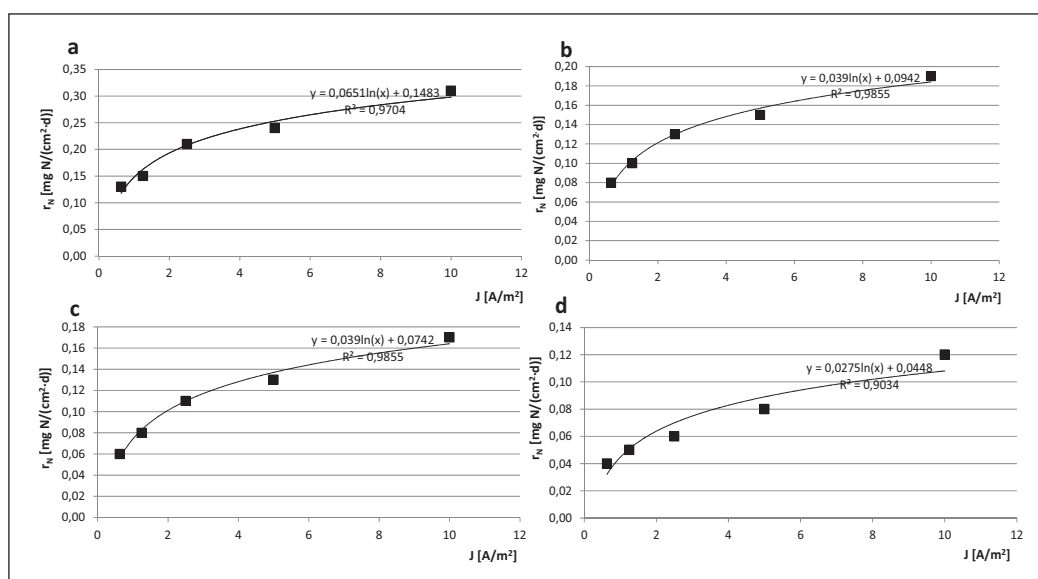


Fig. 3. The rate of nitrogen compounds removal in rotating electrobiological disk contactor depending on the electric current density: a – HRT = 4 h, b – HRT = 8 h, c – HRT = 12 h, d – HRT = 24 h

### Specific rate of denitrification $r_D$ in a rotating electro-biological disk contactor

A correlation was demonstrated between the specific rate of denitrification and electric current density (Fig. 5) when hydraulic retention time ranged from 4 h to 24 h, which was confirmed by high values of the determination coefficient ( $R^2$ ). The  $r_D$  values decreased along with an increasing density of electric current and HRT extension. At HRT = 4 h and  $J = 0.63 \text{ A/m}^2$ , the  $r_D$  value reached  $8.46 \text{ mg N/kWh}$ . The specific rate of denitrification decreased along with an increasing density of the electric current.

Under the flow of electric current with the highest density ( $10.00 \text{ A/m}^2$ ), the  $r_D$  value was  $1.27 \text{ mg N/kWh}$ . Extending hydraulic retention time to HRT = 8 h decreased the specific

rate of denitrification. At the lowest current density ( $J = 0.63 \text{ A/m}^2$ ), the  $r_D$  value was  $5.14 \text{ mg N/kWh}$ . Increasing current density to  $10.00 \text{ A/m}^2$  resulted in the lowest achieved specific rate of nitrogen compounds removal, i.e.,  $r_D = 0.81 \text{ mg N/kWh}$ . Further HRT elongation to 12 h led to a decreased load of total nitrogen removed. In the analyzed range of electric current densities, i.e., from  $0.63 \text{ A/m}^2$  to  $10.00 \text{ A/m}^2$ , the specific rates of denitrification ranged from  $4.35 \text{ mg N/kWh}$  to  $0.74 \text{ mg N/kWh}$ . The lowest  $r_D$  values were determined at HRT = 24 h. At the lowest electric current density of  $0.63 \text{ A/m}^2$ , the  $r_D$  value was the lowest and reached  $2.62 \text{ mg N/kWh}$ , whereas at the highest current density of  $10.00 \text{ A/m}^2$ , it reached  $0.48 \text{ mg N/kWh}$ . The specific rates of denitrification achieved at the other electric current densities, namely,  $1.25 \text{ A/m}^2$ ,

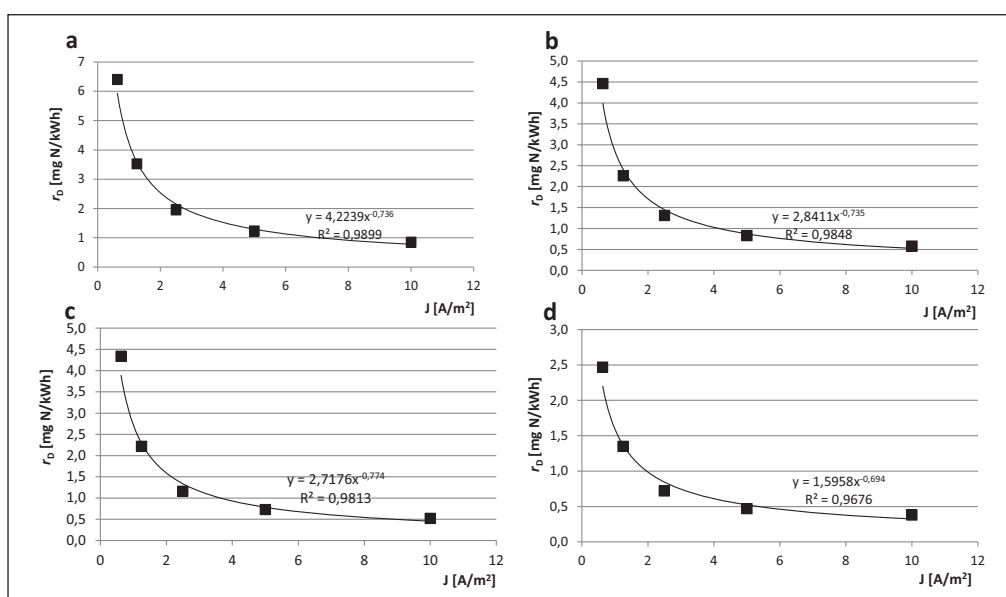


Fig. 4. Specific rate of denitrification in rotating electrochemical disk contactor depending on the electric current density: a – HRT = 4 h, b – HRT = 8 h, c – HRT = 12 h, d – HRT = 24 h

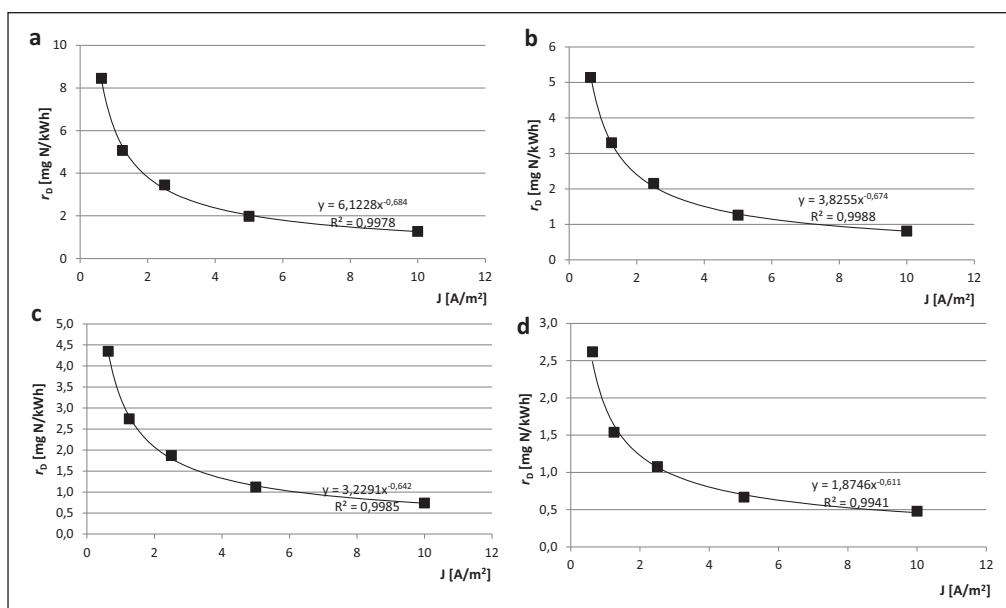


Fig. 5. Specific rate of denitrification in rotating electrobiological disk contactor depending on the electric current density: a – HRT = 4 h, b – HRT = 8 h, c – HRT = 12 h, d – HRT = 24 h

2.50 A/m<sup>2</sup>, and 5.00 A/m<sup>2</sup>, were at 1.54 mg N/kWh, 1.08 mg N/kWh, and 0.67 mg N/kWh, respectively. A comparison of the specific rates of denitrification determined in RECDC and REBDC reactors demonstrates that, at HRT = 4 h, the  $r_D$  value was higher in REBDC than in RECDC, regardless of electric current density. The current density increase led to a growing difference in  $r_D$  values between the reactors. At  $J = 0.63$  A/m<sup>2</sup>, the  $r_D$  was higher by ca. 32% in REBDC than in RECDC (8.46 mg N/kWh and 6.40 mg N/kWh, respectively), whereas at the highest current density, i.e.,  $J = 10.0$  A/m<sup>2</sup>, it was higher by ca. 49% in REBDC than in RECDC (1.27 mg N/kWh and 0.85 mg N/kWh, respectively). Elongation of wastewater hydraulic retention time diminished differences in specific denitrification rates between the electrochemical and electro-biological reactors. At  $J = 0.63$  A/m<sup>2</sup> and HRT = 12 h and HRT = 24 h, the  $r_D$  values were similar, whereas at  $J = 10.0$  A/m<sup>2</sup>, the  $r_D$  value was higher by ca. 26% in REBDC than in RECDC (0.48 and 0.38 mg N/kWh, respectively).

The study showed that higher values of  $r_N$  and  $r_D$  were achieved in the electro-biological contactor. HRT extension diminished the differences in nitrogen removal rates  $r_N$  and specific denitrification rates  $r_D$  between the electrochemical and the electro-biological contactors. From the technological point of view and because of operating costs, the use of REBDC HRT is not justified for long.

In the present study, the rate of nitrates removal ( $r_N$ ) was observed to increase along with an increasing density of electric current and shortening hydraulic retention time. At the lowest current density ( $J = 0.63$  A/m<sup>2</sup>), the removal rate ranged from 0.04 mg N/(cm<sup>2</sup>·d) to 0.13 mg N/(cm<sup>2</sup>·d), whereas at the highest one ( $J = 10.00$  A/m<sup>2</sup>), it ranged from 0.12 mg N/(cm<sup>2</sup>·d) to 0.31 mg N/(cm<sup>2</sup>·d) in the electro-biological contactor (Fig. 3). In the electrochemical contactor, the removal rate ranged from 0.04 mg N/(cm<sup>2</sup>·d) to 0.1 mg N/(cm<sup>2</sup>·d) at the lowest current density ( $J = 0.63$  A/m<sup>2</sup>), as well as from 0.09 mg N/(cm<sup>2</sup>·d) to 0.2 mg N/(cm<sup>2</sup>·d) at the highest one ( $J = 10.00$  A/m<sup>2</sup>) (Fig. 2). These values are higher than those reported by Park et al. (2005), who achieved the maximal nitrates removal rate of 0.17 mg N/(cm<sup>2</sup>·d) at the current intensity of 200 mA ( $J = 19.00$  A/m<sup>2</sup>), and those demonstrated by Sakakibara and Kuroda (1993), who achieved a denitrification rate of 0.038 mg N/(cm<sup>2</sup>·d). Zhou et al. (2007) observed an increase in the effectiveness of nitrates removal and denitrification rate along with an increasing load of inflowing nitrates. This can be explained by an increased number of electron acceptors along with an increasing concentration of nitrates. According to computations of Zhou et al. (2007), the rate of denitrification increased from 0.072 mg N/(cm<sup>2</sup>·d) to 0.222 mg N/(cm<sup>2</sup>·d) ( $J = 0.30$  A/m<sup>2</sup>), when the initial concentration of nitrates increased from 10 mg N/L to 50 mg N/L. When the C:N ratio is lower than the stoichiometric ratio, denitrification is performed mainly with gaseous hydrogen as an electron donor. In the opposite case, nitrates will be removed via hydrogenotrophic and heterotrophic denitrification. Organic matter excess leads to higher denitrification rates and low accumulation and rapid degradation of nitrites (Deng et al. 2016, He et al. 2016). In addition, Zhou et al. (2007) demonstrated that the denitrification rate reached its maximum at the flow rate of 150 cm<sup>3</sup>/h (HRT = 4 h), at which concentrations of both nitrates and nitrites in the effluent met the binding criteria.

This indicates that at HRT < 4 h, the rate of denitrification is limited by the wastewater flow rate and the gaseous hydrogen concentration. A successive increase in the flow rate leads only to incomplete denitrification and accumulation of nitrites. In other research aimed at increasing the effectiveness of the simultaneous removal of nitrates and organic matter, Zhou et al. (2009) achieved a denitrification rate of 0.167 mg N/(cm<sup>2</sup>·d) in a conventional 2D bio-electrochemical reactor at HRT = 4 h ( $J = 0.30$  A/m<sup>2</sup>) and initial nitrates concentration of 30 mg N/L. In turn, in a 3D bio-electrochemical reactor, i.e., in the system with the third bipolar electrode made of activated carbon, the nitrates removal rate was 0.225 mg N/(cm<sup>2</sup>·d) at HRT = 4 h and 0.288 mg N/(cm<sup>2</sup>·d) at HRT = 8 h ( $J = 0.30$  A/m<sup>2</sup>). Sequentially, Vasiliadou et al. (2006) concluded that a higher nitrates concentration (above 40 mg N/L) decreases the rate of hydrogenotrophic denitrification, while nitrites concentration remains low. Such a dependency was, however, not observed in our study.

Considerably lower denitrification rates, compared to the rotating electro-biological disk contactor (0.021 mg N/(cm<sup>2</sup>·d) – 0.87 mg N/(cm<sup>2</sup>·d)), were determined in membrane reactors with a biofilm. In a reactor with a membrane made of polyvinyl chloride (PVC) fibers, Xia et al. (2009) obtained a denitrification rate of 1.2 g N/(m<sup>2</sup>·d) (1.2·10<sup>-4</sup> g N/(cm<sup>2</sup>·d)). Similar results were reported by Lee and Rittmann (2000) –  $r_N = 1.0$  g N/(m<sup>2</sup>·d), Shin et al. (2008) –  $r_N = 1.4$  g N/(m<sup>2</sup>·d), and Terada et al. (2006) – denitrification rate from 3.53 g N/(m<sup>2</sup>·d) to 6.58 g N/(m<sup>2</sup>·d), who studied hydrogenotrophic denitrification in membrane reactors.

Maintaining the appropriate HRT and current intensity in the rotating electro-biological disk contactor creates the required conditions for the development of hydrogenotrophic bacteria, including the optimal concentration of electron donors, favorable pH, and adequate reaction time. Too long HRT leads to process inhibition by the accumulation of nitrites (Zhou et al. 2007). As the current increases, the concentration of hydrogen significantly exceeds the saturation constant, so that excess hydrogen remains in the biofilm and negatively affects the denitrification process (Zhou et al. 2007; 2009). It was confirmed by the research of Rodziewicz et al. (2019), who observed an increasing nitrites concentration with electric current density increase. This research also showed that the pH value of treated wastewater depended on electric current density and hydraulic retention time. The pH value was observed to increase along with increasing values of both these parameters. In turn, Hao et al. (2013) demonstrated a significant impact of HRT on nitrogen removal and nitrite accumulation. They showed that the denitrification rate increased with hydraulic retention time and reached its maximum value at HRT = 10 h.

The specific rate of denitrification depended on electric current density and hydraulic retention time. In the electrochemical reactor, its values increased from 0.38 mg N/kWh (at the highest current density of 10.00 A/m<sup>2</sup> and HRT = 24 h) to 6.4 mg N/kWh (at  $J = 0.63$  A/m<sup>2</sup> and HRT = 4 h). Higher values of the specific rate of denitrification were achieved in the electro-biological contactor. The lowest  $r_D$  value, reaching 0.48 mg N/kWh, was achieved at  $J = 10$  A/m<sup>2</sup> and HRT = 24 h, whereas the highest one, reaching 8.46 mg N/kWh, at  $J = 0.63$  A/m<sup>2</sup> and HRT = 4 h.

However, the above values are lower than those reported by Zhou et al. (2009), who achieved  $r_D$  value of 15.05 g N/kWh at HRT = 4 h in the conventional 2D bio-electrochemical reactor, and even a higher  $r_D$  value in the 3D reactor. At HRT = 4 h, the specific rate of denitrification accounted for 24.37 g N/kWh, whereas at 8 h – for 31.6 g N/kWh. The  $r_D$  values determined at the highest current density of 10.00 A/m<sup>2</sup> and HRT = 24 h are comparable with those reported by Huang et al. (2013).

The specific rate of denitrification decreased when the current density and HRT were increased in the electrochemical and electro-biological rotating contactors. This is because extending HRT and increasing current density in both reactors is associated with greater energy expenditure and higher unit electricity consumption.

## Conclusions

In the electrochemical contactor, a linear dependency was observed between the rate of nitrogen removal ( $r_N$ ) and current density in the examined HRTs, as confirmed by a high value of the determination coefficient  $R^2$ . At HRT = 4 h, the rate of nitrogen compounds removal increased from 0.1 mg N/(cm<sup>2</sup>·d) to 0.2 mg N/(cm<sup>2</sup>·d) at the current density of 0.63 A/m<sup>2</sup> and 10.00 A/m<sup>2</sup>, respectively. At HRT=24 h, this rate increased from 0.04 to 0.09 mg N/(cm<sup>2</sup>·d) at the current densities of 0.63 and 10.00 A/m<sup>2</sup>, respectively.

In the electro-biological contactor, a logarithmic dependency was observed between the rate of total nitrogen removal ( $r_N$ ) and current density in the examined HRTs, as evidenced by a high value of the determination coefficient  $R^2$ . At HRT = 4 h, the rate of total nitrogen removal increased from 0.13 mg N/(cm<sup>2</sup>·d) at  $J = 0.63$  A/m<sup>2</sup> to 0.31 mg N/(cm<sup>2</sup>·d) at  $J = 10.00$  A/m<sup>2</sup>. The lowest rate of total nitrogen removal, reaching 0.04 mg N/(cm<sup>2</sup>·d), was determined in the contactor operated at HRT = 24 h and  $J = 0.63$  A/m<sup>2</sup>.

In both contactors, an exponential dependency was observed between the specific rate of denitrification ( $r_D$ ) and current density during operation at HRTs from 4 h to 24 h. The specific rate of denitrification decreased when the current density and HRT were increased. In the electrochemical contactor operated at HRT = 4 h and  $J = 0.63$  A/m<sup>2</sup>, the specific rate of denitrification was 6.4 mg N/kWh. Increasing current density to 10.00 A/m<sup>2</sup> in this reactor decreased the specific rate of total nitrogen removal to 0.85 mg N/kWh. In the electro-biological contactor operated at the HRT = 4 h and  $J = 0.63$  A/m<sup>2</sup>, the specific rate of denitrification was 8.46 mg N/kWh. Increasing current density to 10.00 A/m<sup>2</sup> in this contactor decreased the specific rate of denitrification to 1.27 mg N/kWh.

The study showed that the rate of total nitrogen removal increased in both electrochemical and electro-biological rotating contactors when the current density was increased and the HRT was decreased.

In both types of contactors, the specific rate of denitrification, which defines the load of nitrogen removed with 1 kWh of electric current, decreased when the current density and HRT were increased. Higher values of  $r_N$  and  $r_D$  were achieved in the electro-biological contactor. HRT extension diminished differences in nitrogen removal rates  $r_N$  and specific denitrification rates  $r_D$  between the electrochemical and the electro-biological contactors.

From the technological point of view and because of operating costs, the use of REBDC is not justified for long HRT.

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## Wpływ gęstości prądu elektrycznego na właściwą szybkość denitryfikacji i na szybkość usuwania azotu w elektrochemicznym i elektrobiologicznym złożu obrotowym

**Streszczenie.** Celem pracy było określenie wpływu gęstości prądu na szybkość usuwania związków azotu ( $r_N$ ) i na specyficzną szybkość procesu denitryfikacji ( $r_D$ ) w elektrochemicznym tarczowym złożu obrotowym (REDC) i elektrobiologicznym tarczowym złożu obrotowym (REBDC). W elektrochemicznym tarczowym złożu obrotowym (REDC) i elektrobiologicznym tarczowym złożu obrotowym (REBDC) katodę stanowiły odpowiednio tarcze pokryte błoną biologiczną i tarcze z których okresowo usuwano błonę biologiczną. Aluminowa anoda była umieszczona w zbiornikach złoża. Badania przeprowadzono na ściekach o wskaźnikach fizykochemicznych podobnych do ścieków z bezglebowej uprawy pomidorów. W pierwszym etapie badań określono wartości  $r_N$  and  $r_D$  w REDC podczas gdy w drugim w REBDC. Zastosowano cztery wartości hydraulicznego czasu zatrzymania (HRT): 4 h, 8 h, 12 h and 24 h dla następujących gęstości prądu: 0.63 A/m<sup>2</sup>, 1.25 A/m<sup>2</sup>, 2.50 A/m<sup>2</sup>, 5.00 A/m<sup>2</sup> and 10.00 A/m<sup>2</sup>. W elektrochemicznym złożu zaobserwowano liniową zależność pomiędzy  $r_N$  i gęstością prądu, podczas gdy w złożu elektrobiologicznym zależność logarytmiczną. Dla obu złożeń stwierdzono wykładniczą zależność pomiędzy  $r_D$  i gęstością prądu. Specyficzną szybkość procesu denitryfikacji malała wraz ze wzrostem gęstości prądu i HRT. Badania pokazały, że w obu złożach, elektrochemicznym i elektrobiologicznym szybkość usuwania związków azotu obniżała się wraz ze wzrostem gęstości prądu i obniżaniem HRT..