Effect of Electrical Current and the External Source of Carbon on the Characteristics of Sludge from the Sequencing Batch Biofilm Reactors

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ABSTRACT
This work presents the results of an experiment on the effect of electrical current density (53, 105, 158 and 210 mA/m²), the type of an external source of carbon (citric acid, potassium bicarbonate) and C/N\textsubscript{NO₃} ratio (0.5, 1.0 and 1.5) on the quantity and quality of formed sludge. The experiment was conducted in sequencing batch biofilm reactors (SBBRs), under anaerobic conditions, with and without the passage of electrical current, under controlled pH of 7.5–8.0. The study demonstrated that in the reactors with electrical current passage and external source of carbon, the volume of sludge increased along with the current density increase from 53 to 158 mA/m². At its highest density (210 mA/m²), the concentration of sludge was insignificantly lower. For all densities of electrical current and C/N\textsubscript{NO₃} values, the concentrations of sludge formed in the reactors with potassium bicarbonate (1.00 to 1.26 g d. m./L) were lower than in the reactors with citric acid (1.26 to 1.30 g d. m./L). The concentration of organic matter was higher in the sludge from the reactors with electrical current passage and potassium bicarbonate, compared to the sludge from the reactors with citric acid. In the reactors with electrical current passage and external source of carbon, the total nitrogen content in the sludge decreased along with the C/N\textsubscript{NO₃} ratio increase for current densities of 53 and 105 mA/m². For a higher electrical current density, the nitrogen content in the sludge was similar. Irrespective of the current density, the nitrogen content in the sludge from the reactors with citric acid was higher than in the sludge from the reactors with potassium bicarbonate. For higher current densities (158 and 210 mA/m²) the increase in the C/N\textsubscript{NO₃} value caused an increase in the P content in the sludge. The electrical current density increase contributed to increasing the content of phosphorus in the sludge. The phosphorus content in the sludge from the reactors with citric acid was lower than in the sludge from the reactors with potassium bicarbonate. The CST values prove that the sludge formed during the wastewater treatment in electrobiological SBBR was characterized by very high dewaterability. The capillary suction time decreased along with increasing the electrical current density but was not significantly affected by the type of carbon source.

Keywords: bio-electrochemical reactor, denitrification, electrocoagulation, citric acid, potassium bicarbonate, sludge

INTRODUCTION
Poland, being poor in natural resources, has to use highly efficient technologies for wastewater treatment to prevent the effects of eutrophication [Janczukowicz and Rodziewicz, 2013; Attour et al., 2014]. It pertains to large, small and household wastewater treatment plants. Pursuant to the regulations binding since the 1st January 2016 stipulated in the regulation of the Minister of Natural Environment of 2014 [Regulations, 2014], the treated wastewater from households or farms discharged to the waters on the area of a municipal agglomeration should meet requirements set for PE agglomeration. This means that the household wastewater treatment plants based on the simplest solutions like a septic tank and pipe draining and located on the area of an ag-
Sludge stabilization is the key process which affects its properties in the perspective of its further management. The most common methods applied to this end in wastewater treatment plants include the aerobic and anaerobic stabilization [Biern et al., 1995; Baran and Turski, 1999]. During the process of stabilization, the organic matter of sludge is mineralized via biochemical transformations. It reduces the sludge demand for oxygen, minimizes the quantity of substances emitting noxious odor, reduces numbers of pathogens, and significantly decreases dry matter content. The American Agency for Environment Protection [Oleszkiewicz, 1999] divides sludge stabilization processes into: (1) those which cause stabilization and partial hygienization of sludge which is then suitable for being injected into the soil or for deposition on a landfill; and (2) those which cause advanced stabilization and hygienization of sludge which may further be used in agriculture (application on soil surface). Oleszkiewicz [1999] postulated adopting the dry matter content decrease by at least 38% as the criterion of a stabilized sludge.

Besides the dry matter content, an equally important parameter of sludge is its susceptibility to dewatering which is determined based on the capillary suction time (CST). The future use of sludge in the natural environment is also largely determined by the presence of biogenes.

Considering the characteristics of wastewater subjected to a two-stage pre-treatment and then treated in a bio-electrochemical reactor as well as the specificity of processes of hydrogenotrophic denitrification and electrocoagulation, the sludge formed in the SBBR as a result of wastewater treatment is expected to have low concentrations of organic compounds, high concentrations of nitrogen and phosphorus, and a short CST. What remains unknown is how the properties of sludge will change as a result of modifying the technological parameters of the process. The available research works provide no data in this respect.

The objective of this study was to determine the effect of the electrical current density, type of the source of organic and inorganic carbon and C/N, ratio on the quantity and quality of the sludge formed in the bio-electrochemical SBBR reactor.

METHODS

The experiments were conducted simultaneously in vertical sequencing batch biofilm reactors (SBBR) with the volume of 3.0 L each (active volume = 2.0 L), under anaerobic conditions (Fig. 1). A set of 12 disks made of stainless steel with diameter of 0.10 m and total surface of 0.19 m² was installed in each reactor. The distance between disks was 5 mm. They were mounted co-axially on a vertical shaft rotating with the speed of 10 rpm; their submersion rate was 100%.
The experiment was conducted under the following conditions: without the passage of electrical current (reactors: R₀, Rₐ, and Rₚₐ), and with the passage of electrical current (reactors: Rₕ₂, Rₐ₊ₕ₂, and Rₚₐ₊ₕ₂). In the control reactor (R₀), without the electrical current flow and without an external source of carbon, the synthetic wastewater was subjected to bio-treatment. In another two reactors (Rₐ and Rₚₐ) without the electrical current flow, citric acid and potassium bicarbonate, respectively, were used in the concentrations ensuring C/N NO₃ ratios of 0.5, 1.0 and 1.5. In the reactors with the passage of electrical current, no external source of carbon was introduced in reactor Rₕ₂, likewise in R₀, whereas citric acid and potassium bicarbonate were fed as carbon sources to reactors Rₐ₊ₕ₂ and Rₚₐ₊ₕ₂, respectively. The wastewater retention time was 24 hours in each reactor.

The reactors were adapted for 3 months to achieve the appropriate structure of the biofilm and stable concentration of nitrogen compounds in the effluent using the activated sludge from denitrification tanks of the Municipal Wastewater Treatment Tank “Łyna” in Olsztyn as the inoculum. The analytical control of the treatment process was begun after the adaptation period.

In the reactors with the electrical current passage, provided by laboratory feeders – Programmable DC Power Supply – HANTEK PPS 2116A – (0–5A) (0–32V) and MANSON DC Power Supply – DPD 3030 (0–3A, 0–30V), disks with immobilized biofilm served as the cathode, whereas an aluminum plate with a total surface area of 0.033 m² served as the anode. The cathode and the anode were connected to the laboratory feeder to ensure the desired density of the electrical current, i.e. 53, 105, 158 and 210 mA/m² (the
current intensity was 10, 20, 30 and 40 mA, the current voltage ranged from 3.0 to 5.0 V). The experiment was conducted for 16 weeks under the conditions of controlled pH (pH 7.5–8.0).

The experiments were conducted with the synthetic wastewater characterized by high concentrations of nitrate nitrogen and total phosphorus, and by a low concentration of organic matter (COD). The parameters of wastewater corresponded to the parameters of municipal sewage subjected to bio-treatment in a municipal wastewater treatment plant with a high efficiency of organic compounds removal and enabling nitrification. The mean composition of wastewater flowing into the reactors was as follows: 50.68 (±1.61) mgN\(_{NO_3}\)/L, 0.0 mgN\(_{NO_2}\)/L, 0.0 mgN\(_{NH_4}\)/L, 5.16 (±0.20) mgP/L, 70.30 (±10) mgO\(_2\)/L, 29.09 mgC/L, InOC – 47.04 mgC/L. The electrolytic conductance of wastewater reached 1.74(±0) mS/cm. The analyses were conducted at a temperature of 25.3(±0)°C.

The analyses of the sludge discharged from reactors included determinations of:
- concentration of sludge with the gravimetric method acc. to PN-EN 12880:2004,
- concentration of total nitrogen in the sludge with the spectrophotometric method based on the procedure developed by Lange LCK company,
- concentration of total phosphorus in the sludge with the spectrophotometric method based on the procedure developed by Lange LCK company,
- COD concentration in the sludge with the spectrophotometric method based on the procedure developed by Lange LCK company,
- capillary suction time (CST) with an electronic CST Meter by ProLabTech, 1112041 CE.

RESULTS AND DISCUSSION

In this experiment, we analyzed the effect of electrical current density, organic and inorganic substrate, and C/N\(_{NO_3}\) ratio on the quantity and quality of the sludge formed in sequencing batch biofilm reactors (SBBRs), under anaerobic conditions with and without the passage of electrical current.

Our earlier investigations demonstrated high efficiencies of the nitrogen and phosphorus compounds removal in the reactors of this type, i.e. SBBR reactors. The efficiency of nitrogen removal increased along with the intensity of electrical current and quantity of carbon fed to the reactor. Higher efficiencies were observed in the reactors fed with citric acid. The highest efficiency was recorded at the current density of 210 mA/m\(^2\) in the reactor with citric acid. At the C/N\(_{NO_3}\) of 0.5, this efficiency accounted for 83.05(±1.16)%, while at C/N\(_{NO_3}\) of 1.5 it reached 87.61(±1.6)%. Similar tendencies were observed in the case of phosphorus. The highest efficiency of its removal reaching 97.69(±2.1)% was determined in the reactor with citric acid used as the source of carbon and at the current density of 210 mA/m\(^2\) and C/N\(_{NO_3}\) ratio of 1.5. The respective value achieved in the reactor with potassium bicarbonate was slightly lower and reached 96.68%.

Literature works emphasize that the volume of the sludge formed in electrochemical reactors depends on the material the electrodes are made of. A study conducted by Lacasa et al. [2011] demonstrated lower production of sludge upon the use of aluminum electrode compared to the Fe electrode. During the analyses carried out at electrical current densities from 10 A/m\(^2\) to 50 A/m\(^2\) and with the AI electrode, the sludge formation ranged from 18.8 g/m\(^2\) to 67.4 g/m\(^2\), whereas with the use of Fe electrode – from 27.4 g/m\(^2\) to 586.4 g/m\(^2\). Significantly higher results were reported by Akyol [2012], who – applying the current density of 35 A/m\(^2\) – determined the sludge concentration of 9.63 kg/m\(^3\) using the Fe electrode and 7.73 kg/m\(^3\) using the AI electrode. According to Gharibi et al. [2013], the concomitant processes of electrolysis and electrocoagulation contribute to the improved sludge dewaterability.

In our experiment, the quantity of sludge (Fig. 2) formed in the reactors without the electrical current passage was insignificantly lower than in the control reactor (R\(_{cp}\), 1.2 g d.m./L). Simultaneously, higher quantities of formed sludge were observed in the R\(_{ca}\) reactors with an external source of organic carbon (citric acid), compared to the R\(_{pb}\) reactors fed with the inorganic carbon (potassium bicarbonate), i.e. from 1.1(±0.09) to 1.2(±0.09) g d.m./L and from 1.008(±0.06) to 1.11(±0.06) g d.m./L, respectively. The lower sludge quantity resulted from the predominance of the processes of heterotrophic denitrification in the reactors with citric acid and the processes of autotrophic denitrification in the reactors with potassium bicarbonate. The mass of autotrophic denitrifiers and, consequently, the quantity of sludge formed as a result of biofilm exfoliation is lower than in the case of heterotrophic denitrifiers [Grady et al., 1999].
In the reactors with the passage of electrical current and with an external source of carbon (R_{CA+H2} and R_{PB+H2}), a tendency could be observed for increasing sludge formation along with the current density increase from 53 to 158 mA/m². The highest concentration of sludge (1.38(±0.03) g d.m./L) was determined in the R_{CA+H2} reactor at C/N_{NO3} of 0.5. At higher current density (210 mA/m²), the sludge concentrations were slightly lower and ranged from 1.26(±0.08) to 1.30(±0.03) g d.m./L. In the case of the reactors with potassium bicarbonate (R_{PB+H2}), the sludge concentrations were lower than in the reactors with citric acid – at all current densities and C/N_{NO3} values and fitted within the range from 1.00 (±0.07) to 1.26 (±0.06) g d.m./L. The concentration of sludge in the reactors with the electrical current passage and an external source of carbon was higher than in the R_{H2} reactor in which electrochemical processes (electrocoagulation and electrochemical reduction of nitrates) and autotrophic denitrification predominated when there was no external source of carbon [Rodziewicz, 2017]. The decreasing concentration of sludge along with the increasing current density observed in R_{H2} (from 1.03(±0.1) to 0.9(±0.11) g d.m./L) prove that the process of organic compounds oxidation to carbon dioxide was more intense at higher densities of the electrical current and at consequently higher temperatures of wastewater. Autotrophic denitrification was expected to predominate in this reactor [Rodziewicz, 2017].

The quantities of the formed sludge (below 1.3 kg/m²) determined in our experiment (fig. 2) were similar to those achieved by Akyol [2012], but many times lower than these reported by this author in the reactor with Al electrode (7.73 kg sludge/m³). They were also significantly lower than the values determined by Rodziewicz [2017] in her study on the treatment of wastewater originating from soilless crop cultivation in an electrobiological disk contactor (3.8 -5.25 kg/m³). This is mainly due to the differences in the quality of wastewater being treated and in the type of carbon source applied.

The lowest COD values (Fig. 3) were determined for the sludge from the control reactor R₀ (13.71(±1.2) mg/g d.m.). This was due to the lowest concentration of organic matter in the wastewater being treated.

The organic compounds present in the analyzed wastewater were consumed mainly by the biomass-forming organisms during the biofilm growth. Part of them was utilized by denitrifying heterotrophs. Feeding an additional source of organic carbon (C₆H₈O₇) and inorganic carbon (KHCO₃) to the reactor increased the content of organic compounds in the sludge. In the sludge from reactors R_{CA} and R_{PB}, the value of COD ranged from 22.17(±1.6) to 23.57(±1.56) mg/g d.m. and from 22.43(±0.1) to 29.41(±0.92) mg/g d.m., respectively, as a result of greater availability of organic compounds in the treated wastewater. In the R_{H2} reactor with the electrical current passage, the content of organic compounds in the sludge was affected by the electrical current den-
The highest content of total nitrogen was determined in the sludge from the control reactor (Fig. 4). Considering the low concentration of organic matter in the wastewater being treated, nitrogen removal occurred mainly as a result of biomass growth. The total nitrogen content in the sludge from R$_0$ reached 12.1(±0.7) mg N/g d.m.

In the reactors without the electrical current passage and external source of carbon, a decrease was noted in the nitrogen content along with the increasing C/N$_{NO_3}$ value, for both types of carbon sources. The nitrogen content was higher (range: 9.5(±0.72) -12.0(±0.22) mg N/g d.m.) in the sludge from the reactors with citric acid (R$_{CA}$) than in the sludge from the reactors with potassium bicarbonate (R$_{PB}$) where it decreased from 10.0(±0.51) to 7.7(±0.45) mg N/g d.m.

The lower contents of nitrogen in this sludge compared to the sludge from R$_0$ confirm that the autotrophic and heterotrophic denitrification did proceed in these reactors though with various intensity [Kłodowska et al., 2014].

In the reactor with the electrical current passage and an external source of carbon (R$_{CA+H_2}$ and R$_{PB+H_2}$), the total nitrogen content in the sludge decreased along with the increasing C/N$_{NO_3}$ value at the current densities of 53 and 105 mA/m$^2$. At the higher densities (158 and 210 mA/m$^2$), the total nitrogen content in the sludge was similar. Regardless of the current density and C/NO$_3$ value, the nitrogen content in the sludge from the reactors with citric acid was higher than in that from the reactors with potassium bicarbonate. In the case of the reactors with citric acid, the highest nitrogen content accounted for 8.2(±0.53) mg N/g d.m. and the lowest one for 5.3(±0.44) mg N/g d.m., whereas in the case of reactors with potassium bicarbonate, the respective values were 7.7(±0.57) mg N/g d.m. and 3.6(±0.31) mg N/g d.m. (fig. 4). These are significantly lower (two, three- and four-fold) from the values typical of the sludge from municipal wastewater treatment plants [Malej, 2000; Determining, 2004], the sludge from the treatment of wastewater from soilless crop cultivation [Rodziewicz, 2017], but two times higher than for the sludge from fish culture [Sikora, 2008]. This is an effect of highly efficient denitrification which proceeded in the

![Fig. 3. Sludge COD values depending on electrical current density and C/N$_{NO_3}$ ratio](image)
reactors with the electrical current passage and of the specific character of the treated wastewater which contained only nitrate nitrogen.

In the case of phosphorus, its low content not exceeding 0.3(±0.02) mg P/g d.m. (Fig. 5) was determined in the sludge from the control reactor. The phosphorus removal in this reactor resulted exclusively from its incorporation into the biomass [Grady et al., 1999]. Introduction of carbon sources to the reactors contributed to a slight increase in the phosphorus concentration (0.4(±0.05) mg P/g d.m.), as a result of the development of biomass with a different characteristics. In contrast, no significant differences were observed in the phosphorus concentrations in the sludge at different C/N\textsubscript{NO}_3 values and different sources of carbon. A more intense phosphorus removal from the wastewater in the electrocoagulation process was already observed as a result of using aluminum electrode in the reactor with electrical current passage. The phosphorus content was higher in the sludge from the reactors with an inorganic source of carbon (R\textsubscript{PB+H}_2) than in the reactors with citric acid (R\textsubscript{CA+H}_2), probably because of the lower quality of sludge. For the current densities 158 and 210 mA/m\textsuperscript{2} a tendency was observed for the P content increase in the sludge along with the C/N\textsubscript{NO}_3 value increasing. Additionally, the electrical current density increase contributed to an increasing content of phosphorus in the sludge formed during the wastewater treatment. This is a commonly known phenomenon described by many scientists. The effectiveness of the electrocoagulation process in removing the phosphorus compounds from wastewater was reported in many works [Tchamango et al., 2010; Behbahani et al., 2013; Shalaby et al., 2014; Kuokkanen et al., 2016; Rodziewicz, 2017]. Kuokkanen et al. [2015], who used Al anode and Fe cathode, achieved a 79% efficiency of total phosphorus removal from the wastewater originating from the mining industry upon the use of electrical current with density of 100 A/m\textsuperscript{2}. Other authors obtained a 98.9% efficiency of synthetic wastewater treatment within 40 minutes [Đuričić et al., 2016].

The phosphorus content determined in the sludge at the highest density of electrical current and the highest C/N\textsubscript{NO}_3 value (1.67(±0.06) mg P/g d.m. (0.17%), Fig. 5) in the reactor with potassium bicarbonate (R\textsubscript{PB+H}_2) is six-fold lower than the values reported by Rodziewicz (2017). In her study, the highest percentage content of phosphorus in sludge dry matter (reaching 0.84%) was obtained at the current density of 10.0 A/m\textsuperscript{2} and HRT of 24 h. It is also six times lower than in the sludge from phosphorus precipitation with aluminum compounds and lower than the values noted for sludge formed during precipitation with lime [Oleszkiewicz, 1998]. The above-mentioned value is also lower than that determined in the sludge from trout culture [Sikora, 2008].

The sludge formed during the wastewater treatment in the electrobiological sequencing batch biofilm reactors was characterized by very high dewaterability. This was indicated by the determined values of capillary suction time (CST),

Fig. 4. Sludge nitrogen content depending on the electrical current density and the C/N\textsubscript{NO}_3 ratio
which did not exceed 10 s for any of the analyzed sludge types (fig. 6). The dehydration rate of the sludge from the reactors with the electrical current passage and an external source of carbon (R\textsubscript{CA+H2} and R\textsubscript{PB+H2}) was higher than that of the sludge from the reactors with the additional carbon source (R\textsubscript{CA} and R\textsubscript{PB}). The longest dewatering time was reported for the sludge from the control reactor R\textsubscript{0} (9.89(±0.54) s).

Worse dewaterability was observed in the case of the sludge from the reactors with citric acid being the source of organic carbon (with the highest CST value reaching 9.96(±0.72) s), whereas a better one for sludge from the reactors with potassium bicarbonate being the source of inorganic carbon (with the lowest CST value reaching 7.76(±0.18) s).

Thus, the use of an inorganic substrate caused a shorter capillary suction time. Similar CST values, accounting for 8.7–9.2 s, were determined for the sludge from the chemical industry conditioned with the Fenton’s reagent [Barbusuński and Filipek, 2000].

The susceptibility of the analyzed sewage sludge to dewatering was also demonstrated by other authors according to whom the CST values for crude municipal sludge were over 300 s [Dębowski et al., 2008]. These authors also showed the feasibility of shortening CST to ca. 50 s by sludge conditioning with the Fenton’s
method. Another work [Piotrowska-Cyplik and Czarnecki, 2005] reported the doses of polyelectrolytes which allowed for shortening the CST. With one of these polyelectrolytes, the capillary suction time was reduced to 14 s.

The results of our experiment prove that the sludge from the SBBR type bio-electrochemical reactor will not require conditioning and will dehydrate easily. Being rich in nutrients, it could be used for environmental purposes. It will pose no hazard to the soil nor to the aquatic environment due to its aluminum compounds, as evidenced earlier by Rodziewicz [2017] who demonstrated that at the electrical current density of 0.63 A/m$^2$ (i.e. higher than in our study), the concentration of aluminum (at HRT= 24 h) was at 0.3 mg/g d. m. sludge.

**CONCLUSIONS**

1. In the reactors with the electrical current passage and an external source of carbon, the volume of the produced sludge increased along with the current density increase from 53 to 158 mA/m$^2$. At its highest density (210 mA/m$^2$), the concentration of sludge was insignificantly lower.

2. For all densities of the electrical current and values of the C/N$_{NO_3}$ ratio, the concentrations of sludge formed in the reactors with potassium bicarbonate were lower than in the reactors with citric acid.

3. The content of organic matter (expressed as COD) was higher in the sludge from the reactors with the electrical current passage and potassium bicarbonate used as the external source of carbon, compared to the sludge from the reactors with citric acid.

4. In the reactors with the electrical current passage and an external source of carbon, the total nitrogen content in the sludge decreased along with the C/N$_{NO_3}$ ratio increase for the current densities of 53 and 105 mA/m$^2$. For higher current densities (158 and 210 mA/m$^2$), the total nitrogen content in the sludge was similar.

5. Irrespecyively of the electrical current density, the nitrogen content in the sludge from the reactors with citric acid was higher than in the sludge from the reactors with potassium bicarbonate.

6. Irrespecyively of the electrical current density, the phosphorus content in the sludge from the reactors with citric acid was lower than in the sludge from the reactors with potassium bicarbonate.

7. For the current densities 158 and 210 mA/m$^2$, the increase in the C/N$_{NO_3}$ ratio caused an increase in the P content in the sludge.

8. The electrical current density increase contributed to increasing the content of phosphorus in the sludge formed during wastewater treatment.

9. The values of capillary suction time (CST) indicate that the sludge formed during the wastewater treatment in the electrobiological sequencing batch reactors with biofilm was characterized by high dewaterability.

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