



## Removal of organic pollutants and surfactants from laundry wastewater in moving bed biofilm reactor technology

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### ABSTRACT

The aim of the research was the testing of the moving bed biofilm reactor (MBBR) pilot plant treatment efficiency of industrial laundry wastewater. The two-stage MBBR pilot plant filled with AnoxKaldnes™ (Lund, Sweden) Z-MBBR carriers (Z-400) was used. The test was carried out on a place in the chosen laundry with the usage of real wastewater. The pilot plant worked in aerobic conditions with a coarse bubble aeration system with daily wastewater flow ( $Q_d$ ) equal to 0.6 m<sup>3</sup>/d (hourly flow  $Q_h = 25$  L/h). The hydraulic retention time was equal to 10.4 h. The concentration of dissolved oxygen in MBBR tanks was maintained on the level of 2–4 mgO<sub>2</sub>/L. The efficiency of chemical oxygen demand removal reached 77%–95%, biochemical oxygen demand (BOD<sub>5</sub>) 88%–96%, sum of anionic and non-ionic surfactants 91%–99%, anionic surfactants 94.8%–99.4% and nonionic surfactants 94.2%–99.5%. After the MBBR start-up period, the treated wastewater quality reached that required by law for wastewater discharged to surface waters. Excluding the periods of phosphorus deficiency, the MBBR installation reached the values of pollutant concentrations permitted by the legal regulations.

*Keywords:* Laundry wastewater; Moving bed biofilm reactor; Surfactants; Organic pollutants; Deficiency of nitrogen; Deficiency of phosphorus

### 1. Introduction

The costs of water acquisition and wastewater discharge to municipal sewer systems are an essential part of the running cost of industrial laundry. Raising fees forced management of the laundry to search for possibilities of reduction of water consumption as well as effective technologies of wastewater treatment. The reduction of water consumption causes an increase in the concentration of wastewater pollutants. Because of that, the laundry investment plans should incorporate the analysis and improvements of water and wastewater management systems including continuous improvement of laundry wastewater treatment.

The quality of laundry wastewater depends on the washing assortment, the amount of water used as well as the type and amounts of used washing agents. Laundry specific pollutants are surfactants [1]. Many methods are used for the treatment of laundry wastewater: physicochemical, chemical (mainly oxidation), biological aerobic and anaerobic [2] as well as combined methods. Several authors also tested membrane processes for laundry wastewater treatment. Positive effects have been reached by the use of coagulation followed by membrane ultrafiltration [3,4]. Coagulation has also been used as a stand-alone process [5]. The total content of surfactants on the level below 2 mg/L, in the outflow wastewater, has been reached by a method based on preliminary coagulation then by flotation followed

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by sand filtration, ozonation and eventually granulated activated carbon filtration [6]. Biological methods are also used for laundry wastewater treatment, for example, membrane bioreactors [7,8], or sequencing batch reactors [1]. The use of a moving bed biofilm reactor (MBBR) gave good results in the treatment of wastewater containing detergents as a stand-alone process [9,10] or as the first stage before laundry water renewal processes [11].

MBBR was developed in Norway in the late 1980 s and early 1990 s [12]. Now, due to advantages of compactness, simplicity, stability and increased reaction rates, it has been successfully employed to treat municipal and industrial wastewater [13] and upgrade existing conventional activated sludge processes [14]. In many respects, MBBR is a flexible alternative to the traditional method of wastewater treatment – an activated sludge method [15]. MBBR incorporates benefits provided by both fixed film and activated sludge processes [16]. In MBBR systems, the biofilm carriers are being mixed inside the reactor and kept in permanent suspension. The system does not require any return sludge recirculation and slowly growing microorganisms, which have become established on the carriers are retained in the system together with the carriers themselves [17]. This allows us to maintain a higher concentration of active biomass in the reactor for biological treatment without increasing the reactor size. Moreover, the attached biomass becomes more specialized [16] and biofilm is more resistant to variation in influent characteristics (e.g., shock loads, pH, temperature, and toxic compounds) [14].

The MBBRs use biofilm carriers of a unique design, to maximize the active biofilm surface area in the reactors (12). The biocarriers differ by dimensions (diameter and height), the available surface area for biofilm development, different sections, that is, parameters affecting the growth of microorganisms, as well as the effectiveness of treatment [14]. That is why one of the research programs on MBBR has involved the study of the improvement of the carrier's properties. Using mathematical modeling, it aims to the optimization of solutions, which means such a construction of a carrier, suiting the conditions in which the carriers will be used [15].

On the other hand, Ødegaard et al. [18] suggest that the shape and size of the carrier do not seem to be significant as long as the effective surface area is the same.

Recently, the material surface properties of MBBR carriers and their modification have also been investigated, in order to enhance the control of microbial attachment and biofilm development as well as MBBR performance by faster reactor startups or increased specific activity per surface area [13]. The most preferred material to produce biocarriers is high-density polyethylene (HDPE). It is due to its plasticity, density, and durability. But high hydrophobicity and low surface energy have been reported to limit initial microbial cell attachment in HDPE carriers [14]. Therefore, to produce carriers, in addition to plastics, other materials are also used, like wood-polymer materials [19].

Also, great emphasis is placed on maintaining a constant, proper thickness of the biofilm on the carriers [14]. The optimal supply of the biofilms with substrates and oxygen is reached at a layer thickness/depth of up to approx. 0.5 mm. As a result, the task to be completed by research

and development is, among others, to create a surface that allows for the growth of biofilms having a maximum thickness of up to 0.5 mm to ensure an appropriate diffusion [17]. In the recently developed "Z" biocarriers optimization of biomass, the increase was achieved by the idea of having instead of openings, a flat surface with a grid of defined height. Biofilm develops in the different "wells" to a certain thickness (e.g., 200  $\mu\text{m}$  in Z-200) controlled by the collision between biocarriers through mixing [14].

Investigated laundry was a large industrial laundry localized in Poland nearby Szczecin city. The laundry washes about 80 tons of linens per day and generates, average daily, ca. 600  $\text{m}^3/\text{d}$  of industrial wastewater. The maximal daily wastewater flow was registered at the level of 800  $\text{m}^3/\text{d}$ . Laundry wastewater may be biodegradable [1].

The aim of the conducted research was an efficiency testing of the MBBR pilot plant with Z-MBBR 400 carriers during the treatment of real industrial laundry wastewater. The test was carried out on a place in the chosen laundry with the usage of real wastewater.

## 2. Materials and methods

### 2.1. Materials and equipment

The MBBR pilot plant was continuously fed by real, mechanically pretreated, averaged industrial laundry wastewater. The used wastewater was a mixture of two wastewater streams. The mainstream (above 90% by volume) was industrial laundry wastewater generated in wet washing processes at the temperatures ranging up to 90°C. The second stream (below 10% by volume), polluted mainly by chlorides, consisted of wastewater from the regeneration processes of ion exchangers used for the softening of the water. The wastewater was mechanically pretreated on a sieve and cooled down to the temperature of ca. 40°C. Cooled down and mechanically pretreated wastewater was used as the MBBR raw wastewater. The wastewater was mainly polluted by impurities washed out from linens as well as washing and auxiliaries agents. Surfactants used in the laundry fulfill the criteria of biodegradation given in the European regulation on detergents (Regulation EC No 648/2004).

The two-stage pilot MBBR filled with AnoxKaldnes™ (Lund, Sweden) Z-MBBR media made of HDPE was used in research. The reactor consisted of two tanks (130 L of active volume each – 260 L total active capacity) was located in real industrial condition on a place in the investigated laundry. The pilot plant worked in aerobic conditions with a coarse bubble aeration system and daily wastewater flow ( $Q_d$ ) equal to 0.6  $\text{m}^3/\text{d}$  (hourly flow  $Q_h = 25 \text{ L/h}$ ). The hydraulic retention time was equal to 10.4 h. The possibility of suspended carriers Z-MBBR 400 (Fig. 1) usage for aerobic treatment of laundry wastewater was tested. The carries Z-MBBR, "saddle" in shape, has a grid on its surface, supporting the biofilm – microorganisms, typical for active sludge and biological bed. The edge of the grid height limited the thickness of the biofilm. The biofilm cannot grow higher than the grid height, and so biofilm thickness was controlled. The carries with the grid height equal to 400  $\mu\text{m}$  were used (Z-400). Carriers were suspended in wastewater and the filling degree was equal to 40% (11.44 kg/130 L).

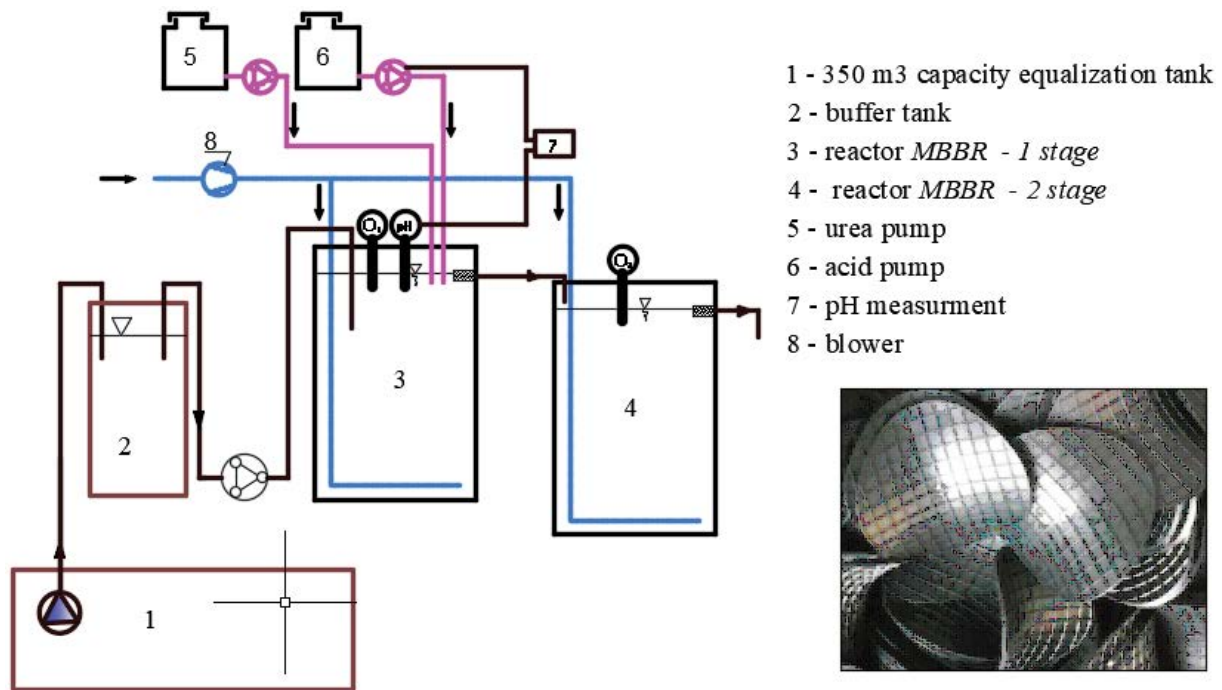


Fig. 1. The technological scheme of MBBR. Photography of AnoxKaldnes™ (Lund, Sweden) Z-MBBR Z-400 carriers.

The scheme of the MBBR pilot plant is presented in Fig. 1.

The MBBR pilot plant was inoculated with biological excess sludge taken from a communal wastewater treatment plant in Gryfino town (Poland) which treats a mixture of communal sewage and laundry wastewater. After inoculation the mixed liquor suspended solids in the MBBR tanks were equal to ca. 1 g/L. The concentration of dissolved oxygen in MBBR tanks was maintained on the level of 2–4 mg O<sub>2</sub>/L. The main tasks for aeration were delivering oxygen necessary for aerobic wastewater treatment and keeping carriers suspended in the wastewater. The biofilm filled the carriers to the height of the grid edges (400 μm) and excess of biofilm was detached by moving carriers colliding and scraping.

To improve a condition of biological wastewater treatment the solution of urea, as a source of nitrogen, was dosing to the first MBBR tank. The commercial Adblue™ solution (Police, Poland) (32.5% w/w of urea content) containing 165.3 g/L of nitrogen was used. The required dose of nitrogen was equal to 5 mg N/L. The pH of wastewater was controlled online. In the case of pH higher than 7.8, the sulphuric acid water solution (H<sub>2</sub>SO<sub>4</sub> ca. 10% w/w) was used for pH correction.

Moreover, the antifoaming agent has been occasionally added to the wastewater.

## 2.2. Analytical methods

Weekly wastewater sampling was done between February and June 2015 (raw, pre-treated wastewater – at the inlet to the first biological chamber of MBBR pilot plant) and between April and June 2015 (treated wastewater – at the outlet from the second chamber of MBBR pilot plant).

The collected samples were used for wastewater quality determination.

The wastewater pH was measured using the HI 991300 portable pH/electrical conductivity/total dissolved solids/temperature meter (Hanna Instruments, Olsztyn, Poland).

Biochemical oxygen demand (BOD<sub>5</sub>) was measured as the pressure difference within a closed system (respirometric BOD – Lovibond BOD, Amesbury, Great Britain – System OxiDirect, Amesbury, Great Britain).

Chemical oxygen demand (COD) was determined by the dichromate method (ISO 6060 standard). Acidified potassium dichromate solution with the addition of mercury sulfate (chloride masking) and sulfuric acid with the addition of silver sulfate as a catalyst was added to the sample of wastewater. The remaining potassium dichromate was titrated with acidified Mohr's salt solution using ferroin as an indicator.

Total phosphorus was determined by the ammonium molybdate spectrometric method (ISO 6878 standard). A sample was mineralized, in a Kjeldahl flask, with sulphuric and then with nitric acids. After cooling down the content of the flask was neutralized with sodium hydroxide solution to pH 3–10. Phosphate ions reacted with molybdate and antimony ions in an acidic solution to form an antimony-phosphomolybdate complex, which was reduced by ascorbic acid to phosphomolybdenum blue. The content of phosphate was measured photometrically at the wavelength of 880 nm (Spectroquant® Pharo 300, Darmstadt, Germany; Perkin Elmer Lambda 20, Waltham, U.S.).

Total nitrogen was determined using the Kjeldahl nitrogen method based on the transformation of nitrogen compounds into ammonium sulfate in the process of sample mineralization with sulfuric acid with the addition of potassium

sulfate and selenium as a catalyst. Ammonia was released from ammonium sulfate by the addition of sodium hydroxide, and distillation to a solution of boric acid/indicator and then the determination of ammonium ion was carried out by titration with hydrochloric acid (EN 25663 standard).

Anionic surfactants were determined as a methylene blue index according to the EN 903 standard. The method is based on the reaction of anionic surfactants with methylene blue to form complexes, which are extracted in chloroform and evaluated photometrically. Surface active agents are concentrated and isolated by gas stripping and the stripped surfactant is dissolved in ethyl acetate. To eliminate interference, the extraction is first from alkaline solution and the extract is then shaken with acidic methylene blue solution.

Non-ionic surfactants were measured according to the ISO 7875–2 standard using Dragendorff reagent. The method, applicable to non-ionic surfactants containing 6–30 alkylene oxide groups, uses gas stripping for surface active agents' concentration. The stripped surfactant is dissolved in ethyl acetate. After phase separation and evaporation of the solvent, the non-ionic surfactant is precipitated in aqueous solution with modified Dragendorff reagent ( $\text{KBiI}_4 \cdot \text{BaCl}_2 \cdot \text{glacial acetic acid}$ ). The precipitate is filtered, washed with glacial acetic acid, and dissolved in ammonium tartrate solution. The bismuth in the solution was titrated potentiometrically (Schott Instruments Titrator TitroLine alpha plus, Mainz, Germany) with pyrrolidinedithiocarbamate solution at pH 4–5 using a bright platinum indicator electrode and a silver/silver chloride reference electrode.

Filtration through glass fiber filters, according to the EN-872 standard was used for suspended solids determination. A sample was filtrated through a glass fiber filter on the vacuum filtration apparatus. The filter was then dried at a temperature of 105°C and the mass of retained solids was determined by the gravimetric method.

The meter HQ40d (Hach, Loveland, U.S.) equipped with an LDO101 sensor was used for dissolved oxygen concentration determination.

In the samples of treated wastewater, the  $\text{BOD}_5$ , COD, total nitrogen, total phosphorus, anionic and non-ionic surfactants were determined.

### 3. Discussion of results

#### 3.1. Raw laundry wastewater quality

The values of determining raw wastewater quality indicators, measured during the MBBR operation, are presented in Table 1.

In the pilot plant test period, the ratio of COD to 5 d ( $\text{COD}_C/\text{BOD}_5$ ) ranged from 1.39 to 2.46. The literature value of the ratio of  $\text{BOD}_5$  to total nitrogen and total phosphorus ( $\text{BOD}_5/\text{N/P}$ ) required for proper wastewater biological treatment, should be equal to 100/5/1 [21]. The median of the  $\text{BOD}_5/\text{N/P}$  ratio in the case of the examined laundry wastewater was equal to 100/2.51/1.37 which indicated a deficiency in nitrogen. The  $\text{BOD}_5/\text{N}$  ratio ranged from 100/1.39 to 100/2.46 and the  $\text{BOD}_5/\text{P}$  ratio ranged from 100/0.28 to 100/6.77. Based on archival data available in the laundry, one can conclude that the deficiency in nitrogen is a characteristic feature of this wastewater. The phosphorus concentration in laundry wastewater was very variable, probably depending on the laundry detergents used. A periodical phosphorus deficiency was observed.

#### 3.2. Quality of the treated laundry wastewater

After 21 d of operation of the MBBR pilot plant, periodic sampling of treated wastewater began. It was assumed that the effect of wastewater treatment would be satisfactory if the quality of treated wastewater is in compliance with legal regulation concerning wastewater dumping to the surface water. The acceptable by law values of the measured indicators are [22]:  $\text{BOD}_5 < 25 \text{ mg/L}$  and  $\text{COD} < 125 \text{ mg/L}$ . Fig. 2 shows the values of COD and  $\text{BOD}_5$  indicators of treated wastewater.

After 30 d of MBBR, pilot plant initialization determined values of  $\text{BOD}_5$  and COD indicators, reached the values of 12–25 and 45–124  $\text{mg O}_2/\text{L}$ , respectively. Exceedances of limit values in two values of the COD indicator and three values of  $\text{BOD}_5$  indicator exceedances were observed. Exceedances were recorded after 1–2 weeks of significant phosphorus deficiency, for  $\text{BOD}_5/\text{P}$  ratio 100/0.35 and 100/0.45. In those cases, the phosphorus content in the wastewater treated was

Table 1  
Quality of the mechanically treated chosen laundry raw wastewater

No.	Parameter/indicator	Allowable limit	Value	
			Maximum	Minimum
1	pH	6.5–9.5	7.9	7.4
2	Chemical oxygen demand, $\text{mgO}_2/\text{L}$	125	1,041	736
3	Biochemical oxygen demand, $\text{mgO}_2/\text{L}$	25	642	347
4	Total suspended solids, $\text{mg/L}$	35	230	140
5	Chlorides, $\text{mg/L}$	1,000	728	238
6	Total phosphorus, $\text{mg P/L}$	2	29.4	1.59
7	Total nitrogen, $\text{mg N/L}$	30	21.8	8.97
8	Anionic surfactants, $\text{mg/L}$	5	17.9 <sup>a</sup>	4.14 <sup>a</sup>
9	Nonionic surfactants, $\text{mg/L}$	10	90.70 <sup>a</sup>	44.80 <sup>a</sup>
10	Conductivity, $\mu\text{S/cm}$		3,592	1,997

<sup>a</sup>Results of the tests are presented in more detail in [20].

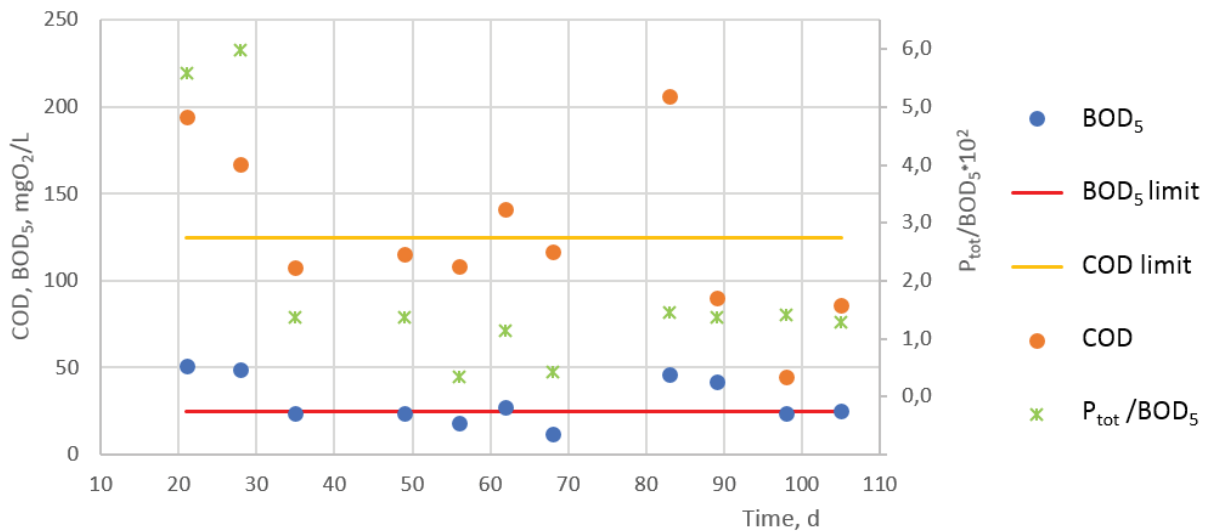


Fig. 2. Treated wastewater COD and BOD<sub>5</sub> indicators values. P<sub>tot</sub> to BOD<sub>5</sub> ratio of the MBBR raw wastewater.

only 35%–45% of the amount necessary for the biological removal of organic compounds.

In Fig. 3 the COD, BOD<sub>5</sub> and surfactant removal efficiencies are presented. The efficiency of BOD<sub>5</sub> removal amounted to 88.4%–98.1% (during the period without exceeding the limit values – 90.5%–98.1%), COD 77.7%–95.1% (during the period without exceeding the limit values – 88.3%–95.1%), anionic surfactants 94.8%–99.4% and non-ionic surfactants 94.2%–99.5%. Excluding the periods of phosphorus deficiency, the MBBR installation reached the values of pollutant concentrations permitted by the regulations [22].

In Fig. 4 the efficiencies of BOD<sub>5</sub> and COD removal related to COD/BOD<sub>5</sub> and total P/BOD<sub>5</sub> ratios are presented.

On the 83rd day of the plant operation, a significant decrease in the efficiencies of BOD<sub>5</sub> and COD removal was observed. At the same time, the highest COD/BOD<sub>5</sub> ratio (2.32) in the wastewater sample was identified. The phosphorus deficiency in the previous sample was also found (Fig. 2).

According to the literature [17], the optimal supply of the biofilms with substrates and oxygen is reached in biofilm up to a thickness of 0.5 mm. In the research, the carriers allowing the growth of the biofilm up to 0.4 mm were used, which should ensure aerobic – anoxic conditions in the whole volume of the biofilm. The observed efficiency of wastewater treatment obtained for the Z-MBBR carriers is was similar to the efficiency obtained using the

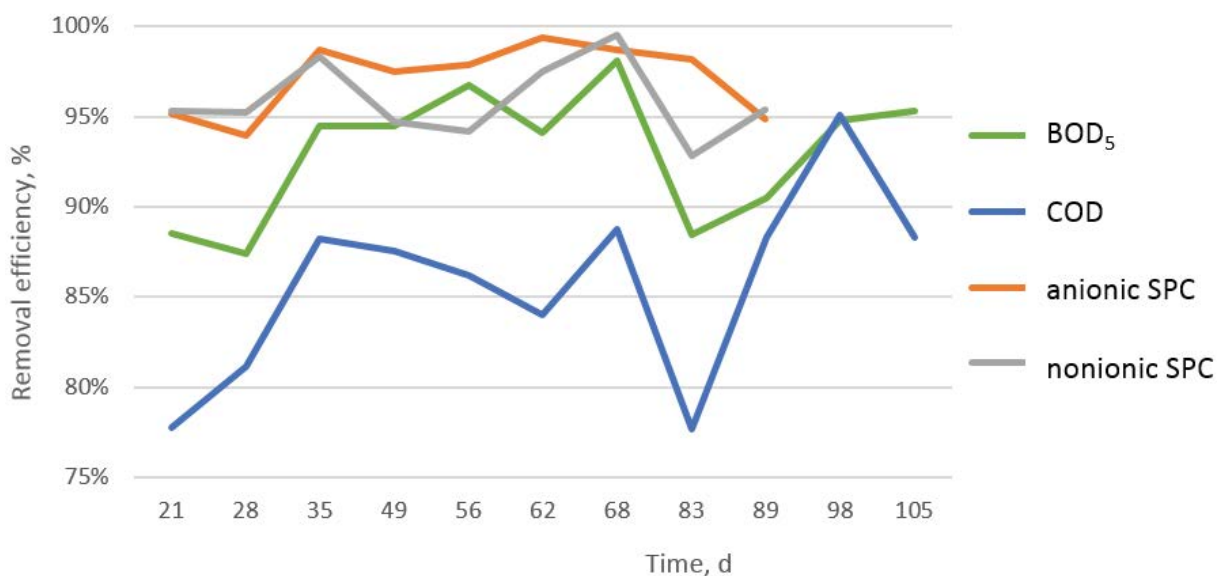


Fig. 3. Laundry wastewater treatment efficiency as reduction of anionic and nonionic surfactant concentration as well as COD and BOD<sub>5</sub> indicators values.

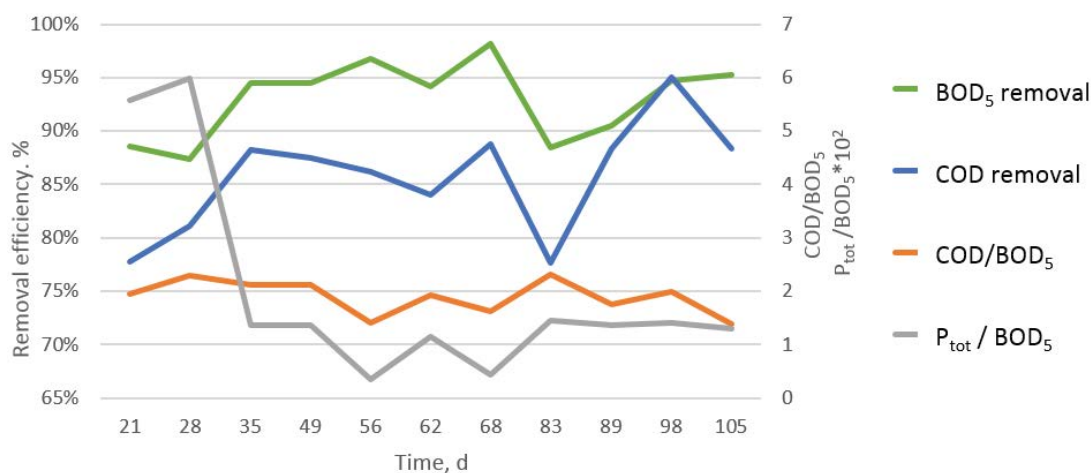


Fig. 4. The efficiencies of BOD<sub>5</sub> and COD removal related to COD/BOD<sub>5</sub> and total P/BOD<sub>5</sub> ratios.

Kaldness type K5 carriers for the same wastewater in the same installation with the same aeration method [23]. For Kaldness K5 carriers removal efficiency of organic pollutants, was equal to 95%–98% for BOD<sub>5</sub> and 89%–94% for COD, and the surfactant removal efficiency was equal to 79%–99% for anionic and 88%–99% for non-ionic ones [23]. Only the removal efficiency of BOD<sub>5</sub> is lower for Z-MBBR carriers than for Kaldness ones. It could have been caused by the periodic occurrence of a significant phosphorus deficit. At the same time, poor excess sludge settleability was observed [24]. The observed COD removal efficiency was similar to that observed by other authors in MBBR [25,26]. The MBBR [26] filled with Z-carriers in 35% volume was applied for the water recycling in the petrochemical industry. The soluble COD removal in the range of 80%–90% was achieved. The surfactant removal efficiency, obtained in the tested MBBR, is comparable to that reached with membrane technology (thermophilic aerobic membrane reactor [27]) used for laundry real wastewater to treatment.

#### 4. Conclusions

- The efficiency of tested MBBR in laundry wastewater treatment, determined as the degree of pollutant removal, was equal to 91%–99% for the sum of anionic and non-ionic surfactants, 88%–96% for BOD<sub>5</sub> and 77%–95% for COD. After the start-up period, the treated wastewater reached the quality required by law for wastewater discharged to surface waters.
- The characteristics of the investigated laundry raw wastewater were a deficiency of nitrogen, wastewater does not require the use of biological nitrogen removal.
- The phosphorus concentration in laundry wastewater was very variable, a periodical phosphorus deficiency was observed. The observed deficit of phosphorus may affect the efficiency of BOD<sub>5</sub> removal.

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