Research on the possibility of using moving bed biofilm reactors for treating car wash wastewater

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ABSTRACT

Wastewater from car washes is an important environmental problem worldwide, both quantitatively and qualitatively. The goal of the article is to investigate the influence of the type and geometry of biofilm carriers on the efficiency of wastewater treatment in moving bed biofilm reactors. The experiments were conducted on laboratory models, in 4 types of biological reactors allowing treatment of 20 L of wastewater each. Three types of biofilter media were used as filling for the bioreactors: Mutag BioChip 30™, BioFLO 9 and Hel-X (H2X36 35/36), and the control group as reference. The tests were carried out for the retention times of wastewater in the bioreactors: 0 h (raw sewage), 1, 3, 6 and 20 h. In each measurement series, the following physical and chemical parameters were measured: organic matter chemical oxygen demand (COD), biochemical oxygen demand (BOD₅), total nitrogen (TN) and total phosphorus (TP), colour, turbidity, pH and conductivity. The conducted experiments proved that the application of filter media (as a biological bed) in each of the tested variants resulted the significant reduction of analyzed wastewater parameters. The main research results indicated Mutag BioChip 30™ carriers as the most effective type of biological filter media. Bioreactors with the aforementioned carriers achieved up to 90% color removal and 94% suspended solids reduction in raw sewage. The COD and BOD₅ reduction were also highly effective, reaching 66%–73% reduction, respectively. The removal of nutrients was also significant, with a reduction of 58% for TN and 68% for TP. The lowest treatment efficiency and circulation parameters were observed for Hel-X (H2X36 35/36) bed.

Keywords: Moving bed biofilm reactors; Biological beds; Car wastewater treatment; Water quality

1. Introduction

Industrial wastewater represents a very broad spectrum of different types of solutions with varying mixtures of pollutants, depending on the nature of the industrial sector in which they were generated [1]. These mixtures differ both in composition and consistency, as well as in toxicity to the aquatic environment, so the conditions for their disposal are subject to strict legal control [2,3]. Car wash wastewater (CWW) is an example of typical industrial wastewater, with varying composition and generation rates. The volume of CWW generated worldwide is an important problem, because they are not recycled with water recovery for the secondary washing process, and the number of functioning treatment plants dedicated only to CWW is negligible [1,4]. The continuous increase in the number of cars in the global market entails a further increase in the number of car washes and the amount of car wash wastewater [4]. This type of wastewater is mostly collected by a sewer and transported to municipal wastewater treatment plants [1].
Due to its chemical composition, CWW can pose a threat to activated sludge technology in wastewater treatment plant bioreactors. Oil and sludge contained in CWW can also coat the pipe walls in sewer systems and cause wastewater flow failures [1]. CWW from many washing stations around the world also contain significant amounts of heavy metals, which are also toxic to activated sludge microbial consortia [4–9]. Considering this possible toxic effects and unfavorable composition of CWW contaminants, classical biological treatment processes are ineffective in a case of this type of wastewater. The main problem in CWW composition is the biochemical oxygen demand (BOD)/chemical oxygen demand (COD) ratio, which is significantly below the critical value of 1/3. This type of wastewater is very difficult to biodegrade and is not suitable for biological treatment processes [6,10–12]. Table 1 presents the concentration of the main pollutants in CWW. These values represent the range of data described in selected scientific publications (as shown in Table 1), however in the real-world conditions their range can be significantly larger [4,13].

The high concentrations of detergents additionally pose risks to the aquatic environment [23,24]. In the scientific sources there is very limited information on industrial CWW treatment by application of biological methods, and the selection of a biological bed for CWW treatment is complicated and requires technological testing to determine the optimal parameters for a biological or hybrid system [1,25,26]. Nadzirah et al. [27], in their review on CWW treatment systems, compiled many methods used in the industry. Despite the various techniques used to treat industrial wastewater, there is a lack of low-cost effective systems to treat CWW efficiently. According to the authors, further advanced research is needed to select the right system to recycle water, with use for washing cars, in a closed circuit. More advanced review studies were conducted by Sarmadi et al. [1] on CWW treatment processes. The authors discussed both existing systems for treating this type of wastewater in the world and laboratory research conducted on wastewater treatment technologies. In the period from 1995 to 2017, they analyzed 332 scientific papers on the industrial wastewater treatment including CWW. Only 36 scientific papers described selected technologies for CWW treatment at laboratory and industrial scales. Their research indicates that there are about 50 wastewater treatment plants in operation worldwide for CWW, using mechanical and physical–chemical processes. Thus, the studies on the efficient and economically justified CWW treatment systems along with water recycling for further washing of vehicles are now essential for the conservation of the world’s water resources and rational use of water [4].

The CWW treatment technology usually includes multi-stage treatment processes with different configurations of types. The initial separation processes employ mechanical sedimentation and flotation methods for oily and non-polar substances. The further stages employ coagulation and flotation methods, advanced oxidation processes, non-woven filters, electrolysis and sand bed filtration, interchangeably. Some systems additionally use a membrane (of various types) purification stage to recycle water in the process of re-washing cars [1,4,23,27–33]. The membrane filtration technologies are also used as independent methods for CWW treatment [34–36], the ultrafiltration and nanofiltration is most commonly used for this purpose [34,35,37], however ultrafiltration can be also combined with reverse osmosis [4,38] or with microfiltration [39]. Unfortunately, these methods are cost-intensive and the research are not implemented in the common CWW treatment process. Biological methods as stand-alone are practically not used in CWW treatment plants, due to the low removing efficiency of selected pollutants. The use of activated sludge technology does not work well in CWW treatment systems due to inappropriate BOD/COD ratios and the presence of toxic substances. Under the conditions of the biological treatment process, the retention time is also significantly increased, which would require the design of large volumes of bioreactor chambers [1,4,28]. Thus, the important scientific challenge is searching for methods to support and/or improve the processes of biological CWW treatment, so that they can be applied in technical conditions to recycle water.

The article describes the results of experiments on the CWW treatment in moving bed biofilm reactors (MBBR), which differed in the type of biological bed used in tested experimental groups. For the tests, the authors chose 3 types of filling media, which varied in the shape and specific surface area of the applied carriers. The control group consisted of bioreactors with activated sludge. The choice of MBBR technology for CWW treatment was due to the high tolerance of this kind of system for the heterogeneity of organic loads and the operation at different temperatures [40].

MBBR bioreactors are currently widely used in the treatment of many types of industrial and municipal wastewater, including those that are difficult to degrade. The selection of biological media, aeration type, and an appropriate hydraulic retention time are crucial in achieving the desired level of pollution reduction in treated wastewater. The carriers media must also be pre-adapted to the conditions and characteristics of the raw wastewater during startup period [41,42].

The study was designed to demonstrate how efficiently CWW can be treated in a period of up to 20 h. The main goal of the research was to investigate the influence of the type and geometry of biofilm carriers on the efficiency of wastewater treatment in moving bed biofilm reactors (MBBR). The conducted research allowed to indicate the possibilities of applying MBBR bioreactors in the process of treating wastewater. In designing technology for the recirculation of water from car wash stations. These studies are crucial in selecting appropriate treatment technologies and configuring

<table>
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<th>Parameter</th>
<th>Value</th>
<th>Literature sources</th>
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<td>[14–19]</td>
</tr>
<tr>
<td>BOD₅, mg·O₂/L</td>
<td>27–650</td>
<td>[2,20,21]</td>
</tr>
<tr>
<td>Turbidity, NTU</td>
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<td>[12,18,19,22]</td>
</tr>
<tr>
<td>Salinity, %</td>
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<tr>
<td>Oil, mg/L</td>
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</table>
selected methods to achieve the desired level of water purification at low process costs.

2. Materials and methods

2.1. Experimental installation

The experiments were conducted in laboratory reactors on real wastewater with a different composition transported from an automatic car wash. The wastewater was averaged (for several days) without mechanical pre-treatment, from a septic tank (collecting wastewater after washing cars from 3 drive-in stands). The experiments were conducted for 3 types of MBBR and a control bioreactor (without moving bed), all tests were realized in parallel in 2 bioreactors for each type of biological reactor. Thus 8 bioreactors were used, each in the shape of polycarbonate cylinder with a height of 1,000 mm and an inner diameter of 194 mm (Fig. 1). At the bottom of each cylinder, an aeration system was installed in the form of an aeration disk (150 mm in diameter and 18 mm high). The disks were connected to the rotameters by Tekalan tubing with an external diameter of 4.3 mm. The flow rate on the rotameter was set at 1 L/min. Such a flow rate, on the one hand, forced adequate movement of the carriers in the bioreactor, on the other hand, did not cause excessive foaming of wastewater. In addition, to minimize foaming, the inlet of the bioreactors was plugged with polyurethane sponges of 10 and 20 PPI gradation.

The reactors operated in batch mode sequencing batch reactor (SBR), in the following arrangements:

- bioreactors A – standard operated SBR with activated sludge, 20 L of wastewater;
- bioreactors B – MBBR reactors, 6 L of Mutag BioChip 30™ media, 20 L of wastewater;
- bioreactors C – MBBR reactors, 10 L of BioFLO 9 media, 20 L of wastewater;

The startup period included 3 weeks of operation of the laboratory system. The biological material used in the experimental bioreactors was an active sludge from the municipal wastewater treatment plant. During the initial period (2 weeks), the bioreactors were filled periodically with CWW and aerated.

2.2. Experiment methods

The experiments were conducted in all reactors in parallel on periodically supplied wastewater. The tests were carried out in 4 large measurement series. Each series was performed on real wastewater with different raw compositions. Each time, experiments were conducted for the same wastewater retention times in the bioreactors: 0 (raw wastewater), 1, 3, 6 and 20 h. The temperature of treated wastewater was 25°C. Wastewater was aerated in normal mode, first 3-h continuous aeration, followed by 1.5-h rigor aeration (nitrification), and 45-min anaerobic phase (denitrification). In each measurement series, a number of the following physical and chemical parameters of wastewater were measured: COD, BOD₅, colour, turbidity, conductivity, pH, total nitrogen (TN), total phosphorus (TP). The research was not focused on analyzing individual organic pollutants or oil-derivatives.

Fig. 1. Scheme of laboratory models for testing car wash wastewater treatment efficiency in bioreactors: A – control, B – Mutag BioChip 30™, C – BioFLO 9, D – Hel-X (H2X36 35/36).
2.3. Types of media used

MBBR, operating in sequencing batch reactor (SBR) mode, was used for the study. Biological methods of wastewater treatment in MBBR mode are based on the technology of biological film, which overgrows the applied media (biofilm carriers of different shapes and an area of active surface). These carriers have a density slightly less than water, so in stagnant water, they float near the surface. In the process of fine bubble aeration, the aqueous solution takes the form of a two-phase fluid (gas–liquid), the density of which is significantly less than water. According to the principles of two-phase fluid mechanics, favorable conditions are created for the circulation of filling media in the reactor. MBBR-type reactors show significantly higher tolerance to temperature changes compared to activated sludge technology and operate stably at temperatures below 10°C. At the laboratory stage of the moving bed biological treatment process, the three most promising types of biomass carriers (biological membranes) were selected (Fig. 2).

The Mutag BioChip 30™ biological bed (Fig. 2A) includes biofilm carriers with high porosity and wavy profile (so that the carriers do not stick together during the treatment process) (Table 2). According to the manufacturer’s recommendations, the optimal filling rate of the reactor is 15%. Depending on the wastewater pollutant load, this rate can be increased to maximum 30%. Bio-media are mainly adapted to the fine bubble type of aeration. The BioFLO 9 biological bed (Fig. 2B) resembles Kaldnes-type media, but its active surface area is significantly larger compared to Kaldnes (Table 2). The slightly lower density of the material causes the carriers to float freely in stagnant water, while in the aerated solution the circulation is very effective, which has a beneficial effect on the wastewater treatment process. With a smaller active surface area compared to Mutag BioChip 30™, the filling level of the carriers in the reactor is in the range of 40%–60%. This type of bio-media is mainly adapted to the fine bubble type of aeration. The Hel-X (H2X36 35/36) biological bed (Fig. 2C) is designed mainly as fixed beds (e.g., as sprinkler beds). Their active surface area (239 m²/m³) allows their use in wastewater treatment processes (Table 2). However, compared to other types of media, their surface area is small. They are mainly used in the treatment of municipal and industrial wastewater from agro-food processing. The filling of the reactor with the bed, due to its small surface area, should be in the range of 50%–70%. With this type of bed, both fine and coarse bubble aeration types can be successfully used during wastewater treatment. The high filling level of the treatment chamber also causes part of the bed to stagnate in the upper layer of the reactor chamber.

2.4. Analytical methods

COD was determined by laboratory dichromate titration method; BOD₅ by the dilution and inoculation method with addition allythiourea reagent; TP with spectrophotometric method with ammonium molybdenum after oxidation of

| Table 2 Parameters of the biological bio-media used for the treatment of car wash wastewater |
|---------------------------------|------------------|-------------------|--------------------------|
| Parameter                       | Mutag BioChip 30™ | BioFLO 9          | Hel-X (H2X36 35/36)     |
| Shape                           | Porous disc with corrugated profile cylinder | Cylinder, outwardly speckled, inside partitions | Cylinder, with tabs to the inside, ribbed in a spiral form |
| Diameter                        | Ø 25 mm           | Ø 9.7 mm          | Ø 35 mm                 |
| Thickness                       | 1.1 mm            | 7 mm              | 36 mm                   |
| Material                        | Pure PE           | Recycled PE and PP| Plastic media with a density close to water |
| Active surface                  | 5,500 m²/m³       | 800 m²/m³         | 239 m²/m³               |
| Colour                          | White             | Black             | Black                   |
peroxide potassium with Cecil CE2021 spectrophotometer; TN with the modified Kjeldahl method with KielFlex K-390 and the potentiometric titration with TitroLine 500; pH and conductivity with the multimeter HQ2100 by Hach Lange, Germany; colour with Cecil CE2021 spectrophotometer; turbidity with Turbidimeter 2100Q by Hach Lange, Germany. Each measurement was performed in the repetition.

2.5. Statistical analysis

The purpose of the study was to test whether time and type of bioreactor would cause changes in the content of individual wastewater quality parameters. Statistical analysis showed no normal distribution, therefore non-parametric Kruskal–Wallis tests were used. When they confirmed statistically significant differences between the results (with \( p \)-value < 0.05), a post-hoc Wilcoxon rank sum test was performed. All statistical analyses were calculated in R software version 4.1.0 with RStudio overlay version 1.4.1717. The level of significance was set as \( \alpha = 0.05 \).

3. Results and discussion

The raw wastewater was characterized by a high degree of heterogeneity of pollutant loads and physical and chemical parameters, which is confirmed by studies by various authors [1,14,23,36]. Table 3 summarizes the averaged values of the tested parameters for the biological treatment process on different biological beds. Due to the high variability of the organics loads and other quality parameters, graphs were used in the interpretation of the results, taking into account the variability of all the parameters tested. The BOD\(_5\)/COD ratio in the CWW used for the process was from 0.01 to 0.68 with mean value 0.16, indicating that they are hardly biodegradable [43].

During the research period, the pollutant loads marked as COD showed variability of up to 280% between the delivered raw sewage (Fig. 3). The differences in the concentrations of readily biodegradable organics were differentiated at the maximum level of 180% (Fig. 3). Sarmadi et al. [1] also reported a wide range of changes in COD values in raw wastewater in their review based on many literature studies. Our results indicate statistically significant changes in the reduction of COD contaminant levels during up to 20 h of the biological treatment process. In addition, the best results were obtained in most cases for variant B, that is, BioMutag moving bed bioreactor (Fig. 3).

It can be concluded that in cases where the salinity level was the highest, not all research groups were characterized by a small biological treatment effect (Fig. 3). Similar relationships were observed by Salehi and Mirbagheri [44] for an activated sludge bed. Tests did not show a reduction of COD to the required level according to the standard [45] in any of the test groups (Fig. 3). Also, studies by other authors indicate that the biological method is not sufficient as an independent CWW treatment system [1,14,23]. In most of the studies and wastewater treatment systems, various authors have presented CWW treatment results on conditioned wastewater [1,23]. The authors of the above article tested raw wastewater without prior treatment, including very high COD and BOD\(_5\) loads.

The concentrations of easily bio-degradable organics were at significantly lower levels compared to COD (Fig. 3). Similar to COD, the differences between baseline concentrations in raw CWW for

<table>
<thead>
<tr>
<th>Time (h)</th>
<th>Sample</th>
<th>COD, mg O(_2)/L</th>
<th>BOD(_5), mg O(_2)/L</th>
<th>TN, mg/L</th>
<th>TP, mg/L</th>
<th>Colour, mg Pt/L</th>
<th>Turbidity, NTU</th>
<th>pH</th>
<th>Conductivity, µS/cm</th>
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<td>180</td>
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<td>7.3</td>
<td>343</td>
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<td>146</td>
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<td>123</td>
<td>552</td>
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<td>109</td>
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<td>104</td>
<td>469</td>
<td>7.6</td>
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<tr>
<td></td>
<td>C</td>
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<td>145</td>
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<td></td>
<td>D</td>
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<td>136</td>
<td>17.1</td>
<td>5.2</td>
<td>260</td>
<td>770</td>
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*Raw wastewater from car wash.
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the treatment process after 20 h were statistically significant for all study groups. The best results were recorded for the bioreactor with the BioMutag bed. Sarmadi et al. [1] in their review paper also presented the results of studies conducted by various authors in which biological treatment processes (mainly on an activated sludge bed) gave partial organic pollutant reduction effects. The treatment process on a biological bed depends mainly on oxygen concentration and mixing. The studied bioreactors were intensively aerated, provided for proper circulation of the biomass carriers in the bed and conditioned oxygen for the processes of biological decomposition of organic matter by microorganisms presented in biofilm on the surface of the bio-carriers. The level of biodegradability of the contaminants determined the efficiency of the process, which was a problem for the studied CWWs due to the very poor BOD/COD ratio.

The variations in total nitrogen concentrations were recorded: up to a maximum of 30.5 mg/L between the raw wastewater samples studied, and up to 13.1 mg/L for total phosphorus concentrations (Fig. 3). Such large changes in phosphorus concentrations were due to the number of detergents used, the main component of which are different forms of phosphates. Statistical analyses showed no statistically significant differences for average values in total nitrogen reduction between the different bioreactors used and little differences in phosphorous values, the best results were observed for BioMutag. The obtained differences are particularly evident for samples with high values of TN (Fig. 3). A similar situation was observed for TP (Fig. 3), where in samples with high values of total phosphorus, differences are seen between concentrations in raw and treated wastewater after 20 h. The overall reduction of nutrients was on average in the range of 27%–63% for TN and 21%–69% for TN during 20 h of process (Fig. 3).

The average concentration of total nitrogen in the delivered car wash wastewater was 23.29 mg/L (17.36–30.50 mg/L). The concentration of nitrogen decreased with increasing time of wastewater retention in bioreactors.

Fig. 3. Changes in car wash wastewater tested parameters: COD and BOD₅ in mg O₂/L; TN and TP in mg/L; turbidity in NTU; colour in mg Pt/l; conductivity in mS/cm, during the treatment process in experimental MBBR: A – control (no bed), B – BioMutag bed, C – BioFLO 9 bed, D – Hel-X bed.
After 1 h of the experiment, the total nitrogen concentration decreased by 26%; after 3 h by 35%, and after 6 and 20 h by 42 and 55%, respectively. Despite the observed differences in nutrient reduction, the tested bioreactors were not able to effectively remove the analyzed forms of nutrients. In published research papers, the biological methods employed in laboratory models also did not determine the efficient removal of nutrients from this type of wastewater [1,14,23]. Further advanced research is needed on modifying bioreactor operating terms to improve the biological removal of TN and TP.

The turbidity and colour of raw wastewater strongly correlate with COD concentration and show similar variability in the collected samples. The level of turbidity concentration between different batches of raw wastewater was up to a maximum of 2,200%, and in a case colour – up to about 1,100% (Fig. 3). Such significant changes are influenced by atmospheric conditions and the amount of inorganic matter that, along with road mud, is washed off the bodies of cars at car washes.

During the experiment, the colour reduction was very evident for all bioreactors between 3–20 h of the treatment process and decreased by an average of 78% after 3 h, and by 87% and 90% after 6 h and 20 h, respectively (Fig. 3). The average turbidity of the raw wastewater was 1,084 NTU (193–3,309 NTU). Turbidity decreased by 41% after 1 h of bed operation, by 63% after 3 h, by 86% after 6 h, and by as much as 94% after 20 h (Fig. 3). The minimum turbidity value was observed after 20 h of the experiment in bed B. Changes from 3 h onward were statistically significant.

Statistical tests confirmed statistically significant differences in the tested parameters tested and between the tested bioreactors (Fig. 3). Post-hoc Wilcoxon tests similarly to other parameters showed differences in results for the operation of the tested bioreactors both colour and turbidity (mainly after 20 h of purification). The best results were seen for the BioMutag moving bed bioreactors.

Despite changes in other physical and chemical parameters, conductivity showed no great differences that did not affect the parameters of the treatment process (Fig. 3). The study was conducted in winter, which is why the differences in conductivity were so drastic. The slush salt washed off at the car washes was the main reason for the differences in conductivity values, up to a maximum of 520% between the raw wastewater samples tested (Fig. 3). After the winter period from April onward, the salt concentration decreases significantly, which translated into conductivity values that were in the range of 700–1,900 mS/cm. There were changes in pH values during 20 h of the treatment process in all test bioreactors A–D (Fig. 3). It is a normal effect of long aeration.

The biological treatment process does not significantly affect both the physical and chemical parameters. There were also no statistically significant differences in pH and conductivity changes between raw and treated wastewater during the treatment process, as well as between the bioreactors tested (Table 2).

The CWW treatment process in the laboratory model of MBBR bioreactors in batch mode generated statistically significant differences between results for different times of sewage retention. Determining the optimal retention time plays a key role in modeling the treatment process in bioreactors of this type [40]. For easily-biodegradable wastewater, these times can generally be shortened [46]; unfortunately, difficult-biodegradable wastewater requires a significantly longer treatment process [47]. The above correlations were confirmed in the authors’ results, in the studied times of 1, 3, 6 and 20 h effective reduction were obtained in most cases after 20 h of the process. Shorter periods did not give satisfactory results. Unfortunately for CWW, even the 20-h retention time was not long enough to achieve a treatment effect, allowing water to be recycled for secondary car washing without further treatment. Longer wastewater retention times are associated with the need to build bioreactor chambers with large operating volumes [48,49].

Operated BioChip biomass carriers confirmed their effectiveness in treating raw CWW. The results for most of the studied parameters were the best for bioreactor B (Fig. 3). Many authors in their studies have shown similar relationships for this type of deposit [50,51].

Operation of the bioreactors was carried out in fine bubble aeration mode, which was able to ensure proper circulation of biomedia carriers in the bed and adequate oxygen concentration above 2 mg/L [52]. Previous unpublished studies by the authors indicated that more efficient micro-aeration (for necessary oxygen transfer into solution) is not able to force proper circulation of the moving bed. The changes of the air flow level can significantly improve or impair CWW treatment parameters. Excessive flow increases the collision of fittings with each other and affects excessive biofilm loss (in the self-cleaning process) [53,54]. In the conducted studies, full circulation of the bed was observed in all tested variants. An air flow is one of the process parameters which can be optimized in this type of industrial wastewater treatment plant [55]. In the experiments, the flow rate was experimentally determined to be 1.5 L/min, which for the used stone diffusers conditioned the necessary O₂ concentration and maintained full circulation without biofilm losses on the surface of the fittings.

4. Conclusions

The BOD₅/COD ratio in the tested raw wastewater was significantly below the 1/3 value, making it hardly biodegradable. Bioreactors operating in mobile biological bed mode show a high level of reduction of pollutants presented in the studied wastewater, especially BOD₅, COD, colour, total nitrogen and phosphorus. Unfortunately, without the support of chemical coagulation, adsorption and mechanical filtration they are not able to effectively treat wastewater to the desired quality within 20 h. Among the tested biological beds, the BioMutag biofilm carriers showed the best reduction effects. The other beds, BioFLO 9 and Hel-X, brought a significant level of reduction in most of the tested wastewater parameters compared to the control group, but worse than BioMutag. The results of the study and the scientific premises allow for the selection of the BioMutag carriers as suitable for the biological treatment of CWW in a technological system that includes the stages of mechanical and physical–chemical wastewater treatment. Biological treatment of car wash wastewater requires preliminary methods that allow these raw wastewaters to be pretreated into an easily biodegradable mixture.
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References


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