

Biological Treatment of Post-Nanofiltration Concentrate of Real Textile Wastewater

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Abstract

The paper evaluates the different bioreactor systems in the treatment of real textile wastewater containing Reactive Red 120 dye concentrated in nanofiltration processes as well as real textile wastewater taken from a working factory. The two-stage sequential semi-fed batch bioreactors and the continuous system with activated carbon used as support material in the anaerobic fixed film bioreactor combined with an aerobic bioreactor, after an acclimatisation period, were very efficient in colour removal, which achieved 95%. The aromatic amine (orphanic acid) released as a result of the azo bond cleavage was traced both in anaerobic (accumulation) and aerobic (degradation) conditions. Biogas production in both systems was very low. Nevertheless indirect biogas production assessment showed some biogas production potential in nanofiltration concentrate without an additional carbon source. Furthermore the nanofiltration concentrate did not inhibit biogas production from synthetic wastewater. Scanning electron microscope visualisation allowed for investigation of biofilm on activated carbon which consisted of at least two layers.

Key words: azo dye biodegradation, nanofiltration concentrate, textile wastewater, biofilm, biogas.

Introduction

Nowadays European law insists on environmentally sustainable development aiming at the preservation of clean water resources in all industrial sectors [1]. The textile industry demands huge quantities of water and a wide spectrum of chemicals. Typically the production of 1 kg of coloured fabric can generate from 70 to 400 l highly loaded wastewater with chemical compounds (i.e. dyes, detergents etc.) [2].

Literature provides a variety of different chemical and physical methods which have been applied to textile wastewater treatment. Techniques such as ozonation, advanced oxidation processes, Fenton reagents, electrochemical destruction, photodestruction or adsorption (for example, on activated carbon) have proven their potential for decolourisation [3 - 5]. Although they are very efficient on a small lab-scale their application on a large-scale can be limited by their costs or huge sludge generation (Fenton reagents, adsorption techniques).

Nanofiltration is a well-known and efficient method for the separation of pollutants from water, which allows for closing the technological water cycle in dye-houses. Qin et al. [6] concluded that nanofiltration allowed 70% of water to recover from effluents. However, a high concentration of dyes and other chemicals in the concentrate (retentate) remaining after the nanofiltration process poses a serious problem. There are

only a few examples of the treatment of the nanofiltration concentrate of textile wastewater. The two-stage-nanofiltration process proposed by Van der Bruggen et al. [7] makes possible the direct reuse of permeate and energy recovery during the incineration of brine concentrated in membrane distillation. Bechtold and Turcanu [8] presented the direct cathodic reduction of azo-dyes in nanofiltration concentrates. Nevertheless the methods mentioned above might be characterized by high energy consumption. Biological methods are generally regarded as environmentally friendly. Furthermore they can lead to the complete mineralisation of organic pollutants at a relatively low cost [9]. A number of available papers on azo dye biodegradation indicate that it is still a very important issue [10 - 12]. Azo dyes represent the largest class of colorants and are the most commonly used in the textile industry (around 60 - 70%). Their presence in textile industry wastewater causes not only a negative aesthetic problem, a decrease in water transparency and lower gas solubility connected with the presence of dyes, but also the potential toxicity and carcinogenic effect of dye decomposition, i.e. aromatic amines. It should be mentioned that azo bonds are generally regarded as not easily biodegradable in conventional aerobic processes. Therefore the ultimate biodegradation of azo-dyes requires a combination of two stages, an anaerobic reduction of the azo bond and aerobic oxidation of the aromatic amines formed. The main shortcoming of the anaerobic reduction processes of azo dyes is the rather slow

reaction rate, resulting in long hydraulic retention times. In order to overcome this problem, Van der Zee et al. [13] utilised the catalytic properties of activated carbon, applying it as a regenerable redox mediator in anaerobic bioreactors. Furthermore Barragán et al. [14] concluded that activated carbon, in comparison to kaolin or bentonite, provided the best conditions for microbial growth.

The decolourisation of azo-dyes was thoroughly investigated but there is limited knowledge of nanofiltration concentrate biological treatment. Anoxic degradation of azo-dyes was applied by Żyła et al. [15] to treat nanofiltration concentrate. The successful application of an up-flow anaerobic sludge blanket (UASB) bioreactor for colour removal from nanofiltration concentration was found in the work by Gomes et al. [16]. The above-mentioned papers did not concern the aromatic amine degradation. The aim of this study was to evaluate the applicability of two-stage sequential batch bioreactors An/Ae and a continuous system consisting of a fixed film bioreactor combined with an aerobic continuous stirred bioreactor (FFB/CSB) to highly concentrated real textile wastewater containing Reactive Red 120 dye generated in nanofiltration processes, as well as real textile wastewater taken from a working dyehouse. Biogas production in the systems mentioned above was investigated. Furthermore indirect biogas production assessment was applied to the nanofiltration concentrate and real textile wastewater. The development and structure of the

biofilm generated on the activated carbon surface was investigated on the basis of scanning electron microscope visualisation. The results presented in this paper demonstrate the coupling of physical and biological methods for textile wastewater treatment. The experiments were focused not only on decolourisation but also on aromatic amine degradation and COD reduction. The results presented may be used as a basis for a pilot plant installation in a dye-house near Lodz (Poland).

■ Experimental

Nanofiltration concentrate of textile wastewater

Textile wastewater coming from the dyeing process (followed by washing and rinsing) of the knitted cotton fabric with reactive dye C.I. Reactive Red 120 (RR 120 – **Figure 1**, $\lambda_{\max} = 512 \text{ nm}$) in a Pyrotec S laboratory dyeing machine (Roaches, UK), according to procedures recommended for this dye, was nanofiltered with a DL polymer membrane (TF polymer, reaction size 96 MgSO_4 , 25°C , pH range 2 - 11, typical flux/psi 31/100). For the sake of the studies presented, a two-fold concentration degree was achieved, which means that 50% of the total volume was collected as a permeate. More detailed information on the experimental set-up for nanofiltration can be found in the work by Sójka - Ledakowicz et al. [17]. The initial BOD/COD ratio in the nanofiltration concentrate was 0.07. Thus in all experiments the nanofiltration concentrate was supplemented with 10% of a concentrated synthetic wastewater solution of the following composition all in $\text{g}\cdot\text{dm}^{-3}$: glucose - 20, acetic acid - 5, casein peptone - 1.56, dry broth - 1.05, NH_4Cl - 0.20, NaCl - 0.07, $\text{CaCl}_2\cdot 6\text{H}_2\text{O}$ - 0.075, $\text{MgSO}_4\cdot 7\text{H}_2\text{O}$ - 0.02, KH_2PO_4 - 0.20, K_2HPO_4 - 0.50. Sulphuric acid of pure grade was used as a neutralising agent.

Real textile wastewater

Real textile wastewater coming from all technological processes was taken from an operating dye house. Wastewater was taken three times (I, II, III) at one week intervals, which provided diversification in its composition due to the application of different dyes and dyeing procedures. The initial BOD/COD ratio for I, II & III was, respectively, 0.29, 0.18, and 0.23, which may suggest the need for the supplementation of wastewater II with biodegradable carbon compounds, which was further confirmed during experi-

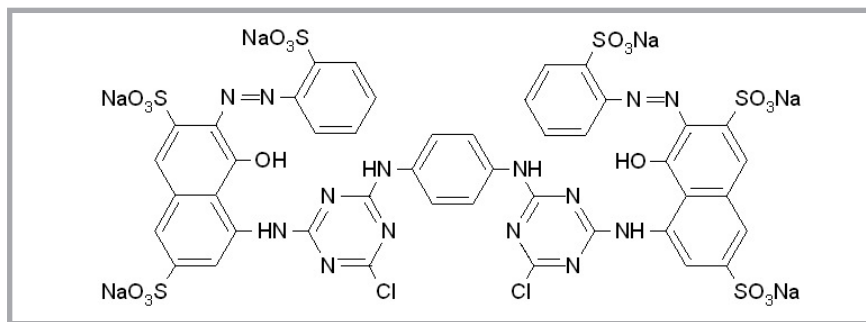


Figure 1. Chemical structure of Reactive Red 120 (as sodium salt).

ments. The pH of real textile wastewater was around 8.8 and was not regulated.

Analytical methods

Two parameters were determined in the bioreactors: pH (WTW pH-meter, Germany) and redox potential (using SenTix ORP electrode). Samples were collected from bioreactors at regular 24 h time intervals, and after centrifugation the following parameters were determined: chemical oxygen demand (COD standard dichromate method, Hach), concentration of orthanilic acid (aromatic amine realised from the RR120 molecule – HPLC analysis as described in work [18]), ad colour - spectrophotometric analysis (Spectrolab UV-VIS spectrophotometer). Samples were scanned in the range of $\lambda = 400 - 800 \text{ nm}$ to observe the shifting of peaks due to dye transformations after the biological treatment. Additionally as only one dye was used in the dye bath in the case of nanofiltration concentrate, the absorbance was measured for one wavelength – 512 nm, where the maximum absorbance for RR120 occurs.

The real textile wastewater contained unknown dyes since the *DFZ* parameter was calculated in accordance with DIN – 38404/1 standards at three wavelengths 436, 525, 620:

$$DFZ = \frac{1000 \cdot E(\lambda)}{d}, \text{ in m}^{-1}$$

where, $E(\lambda)$ – absorbance for a given wavelength, d – thickness of the absorption cell in mm.

Both bioreactors systems examined had biogas collecting and volume measurement systems (liquid displacement method). Furthermore the biogas production potential of real textile wastewater and nanofiltration concentrate without an additional carbon source was determined. Moreover nanofiltration concentrate and water as a control were supplemented

with 10% of synthetic wastewater in order to check whether nanofiltration concentrate inhibits biogas production. OxiTop bottles (Self-check Measurement WTW) were filled with 200 ml of anaerobically digested sludge and 200 ml of samples (control 200 ml of water), where the pH was adjusted to 7. After 13 days, pressure changes in the bottles were compared, which allowed for indirect biogas production assessment.

Experimental set-up

The two-stage system consisted of an anaerobic bioreactor (temperature $37 \pm 1^\circ\text{C}$, working volume 0.4 dm^3 , hydraulic retention time HRT 48 h, the sludge retention time SRT was longer than 21 days - sludge was not drawn from the bioreactors, mixing speed 100 rpm), aerobic bioreactor (operating at an ambient air temperature of $20 - 25^\circ\text{C}$, working volume 0.8 dm^3 , HRT 96 h, SRT longer than 21 days, aeration rate – 0.8 vvm) and a transition tank. The system of peristaltic pumps and digital timers enabled the filling and drawing of all bioreactors and the transition tank. The aerobic reactor was inoculated with activated sludge (SAS), while the anaerobic - with anaerobically digested sludge (ADS), both of which were taken from the municipal wastewater treatment plant. The system worked as a semi fed batch in the 24 h cycle. More detailed information on the above-mentioned system can be found in work [18].

The continuous system consisted of a fixed film bioreactor (FFB) with biomass immobilised on activated carbon (DG 1 - 3 + S, Rütgers Carbo Tech GmbH), and the aerobic was continuously stirred by the bioreactor (CSB) with an overflow and clarifier. The FFB working volume was 450 ml and the flow rate $420 \text{ ml}\cdot\text{d}^{-1}$. In order to immobilise microorganisms, a portion of activated carbon was submerged in ADS for one month. During

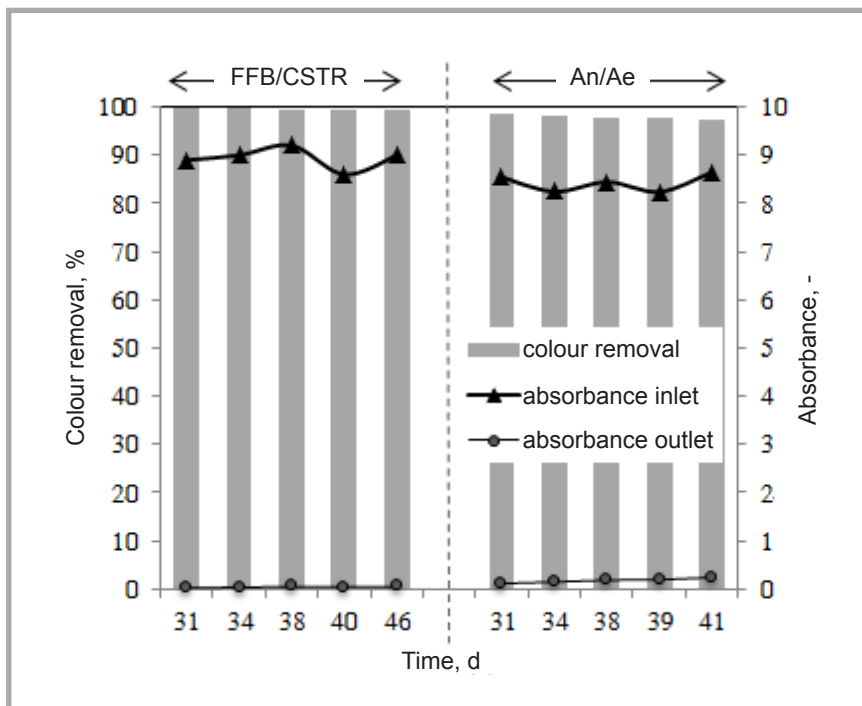


Figure 2. Colour removal from nanofiltration concentrate in FFB/CSTR reactor system and An/Ae.

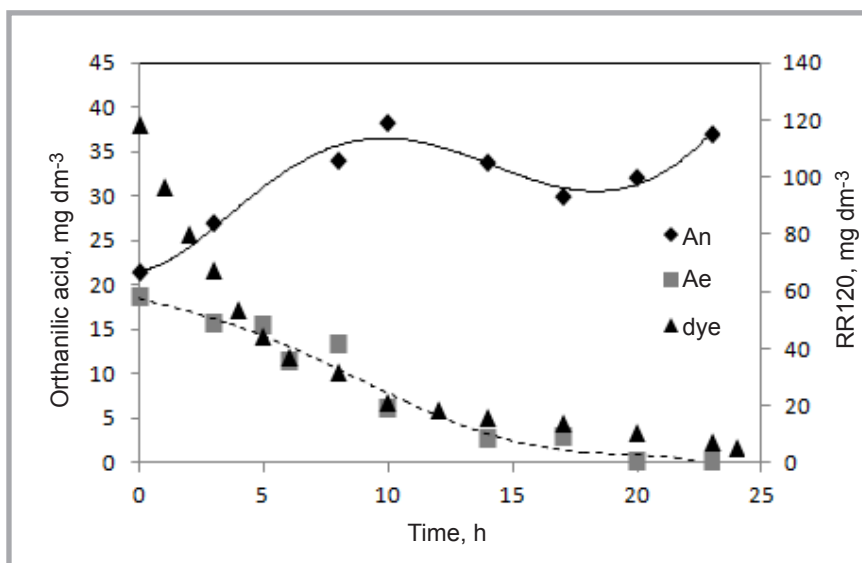


Figure 3. Orthonilic acid concentration in one cycle of An/Ae.

the incubation period, microorganisms were fed with the synthetic wastewater solution. Afterwards non-fixed microorganism and other solid particles were washed out and activated carbon was placed into a glass bioreactor with a perforated bottom, which kept the bed in a desired position [19]. The aerobic reactor was inoculated with SAS. In order to assess the activated carbon capacity to adsorb RR 120 from concentrate and orthonilic acid from its solution, sorption experiments were conducted by the ordinary immersion method [19].

Scanning electron microscope observations of activated carbon surface

After two months of operation, the surface of activated carbon taken from the FFB was visualised by scanning electron microscope (SEM) to observe the colonisation of microorganisms. The surface of the activated carbon prior to and after inoculation (of the working bioreactor) was scanned with a scanning electron microscope - FEI Quanta 200F (USA), which allows for analysis of biological samples without dewatering. The process

was carried out under a low vacuum of 100 Pa. The samples were not pre-treated in any way. Both pure activated carbon and carbon derived from the column were placed in sterile containers with the liquid medium used in bioreactor studies. Grains of activated carbon were transferred to microscope tables covered with carbon tape. The part of particles of activated carbon collected was washed out with a low pressure water stream in order to remove the biofilm.

Results and discussion

Nanofiltration of textile wastewater

It was found that the best results of nanofiltration were obtained at pH range 7 - 10, temperature below 60 °C and pressure 1.5 MPa. An over 90% dye hold-up was obtained for all dye baths. The most favourable conditions of nanofiltration were obtained in the case of a mixture of all technological streams [17]. Filtration after nanofiltration was used as technical water in dyeing processes, whereas retentate was degraded by means of anaerobic azo dye reduction and aerobic oxidation of released aromatic amine.

Colour removal

The nanofiltration concentrate was heavily loaded with electrolytes as a result of the dyeing procedure (conductivity 16 mS·cm⁻¹). Despite the high concentration of chemical substances, the adaptation of the microbial community towards the toxic or recalcitrant compounds by gradual increasing of the textile wastewater concentrate ratio in the inflow (first 30 days of process) allowed to achieve a satisfying decolourisation degree higher than 95% for the semi fed batch bioreactors and 99% in the case of the continuous system (Figure 2). Both systems' performance was monitored for the next 30 days. Colour removal was stable, and the absorbance of the inlet in both systems was around 9, while the outlet in the case of An/Ae was lower than 0.1 and for FFB/CSB lower than 0.05.

On the basis of the assessed adsorption capacity of the AC used, it was confirmed that after saturation with the dye, the colour was eliminated by means of the biological processes [19]. However, activated carbon has great adsorption capacity for the aromatic amine investigated and the concentration of orthonilic acid in the outflow from FFB was negligible. Therefore the capability of ADS used as inocu-

lum to degrade azo bonds was confirmed by kinetic analysis performed for a one operation cycle of An/Ae. Simultaneously with a decrease in dye concentration in the An bioreactor, the concentration of t orthanilic acid increased (*Figure 3*). However, the amount of aromatic amine yielded from the amount of dye delivered did not satisfy the balance, probably due to the adsorption of the amine onto the sludge. According to Ong et al. [20], aromatic amines can also be partially metabolised to generate reductive equivalents for the reduction of the azo bond. The capability of SAS to degrade the aromatic amine was confirmed on the basis of kinetic analysis of the Ae bioreactor's performance. The aromatic amine investigated was completely degraded in aerobic conditions in the presence of SAS (*Figure 3*).

As was shown above, the FFB was very efficient in nanofiltration concentrate decolourisation. Furthermore it also allowed for continuous and efficient decolourisation of real textile wastewater containing unknown dyes. Mean colour removal in the case of real textile wastewater samples I and II, calculated on the basis of DFZ, was 81% in 436 nm, 90% in 525 nm and 94% in 620 nm. In the case of real textile wastewater II, it was lower by 67, 77 and 87%, respectively. Due to differences in colour, dyed fabric and dyeing procedures, the real textile wastewater composition can vary significantly, affecting the efficiency and performance of the treatment method. The first and third sample of wastewater had a sufficient amount of easily biodegradable co-substrate and, contrary to the second sample, were decolourised without an additional carbon source (*Figure 4*).

Carbon metabolism

COD removal in both cases was high - around 90.0% (*Figure 5*). However, effluents after both systems exceeded the legally approved level slightly- 125 mg COD·dm⁻³ [21]. The nanofiltration concentrate of textile wastewater was a strongly alkaline solution (pH above 10). Although, neutralizing agents were used, the average pH in the bioreactors was not stable and had a tendency to increase even to pH 9 in the aerobic bioreactors, which was above the optimal range for the SAS and ADS. However, such a high pH level did not inhibit decolourisation or COD removal. Modi et al. [22] reported that the optimum pH for decolourisation of

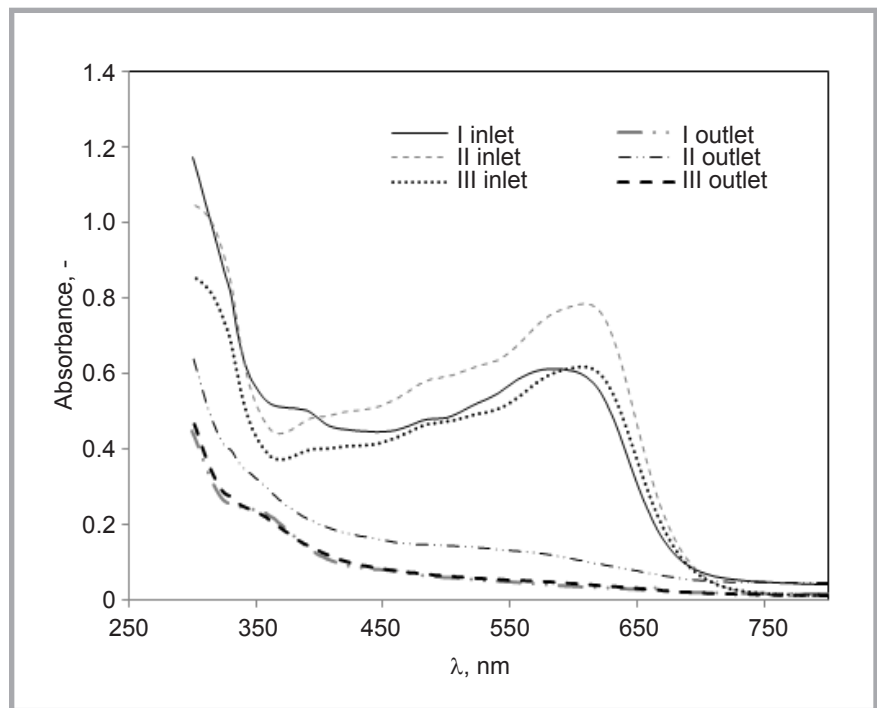


Figure 4. Visible absorption spectrum of real textile wastewater before and after biological treatment.

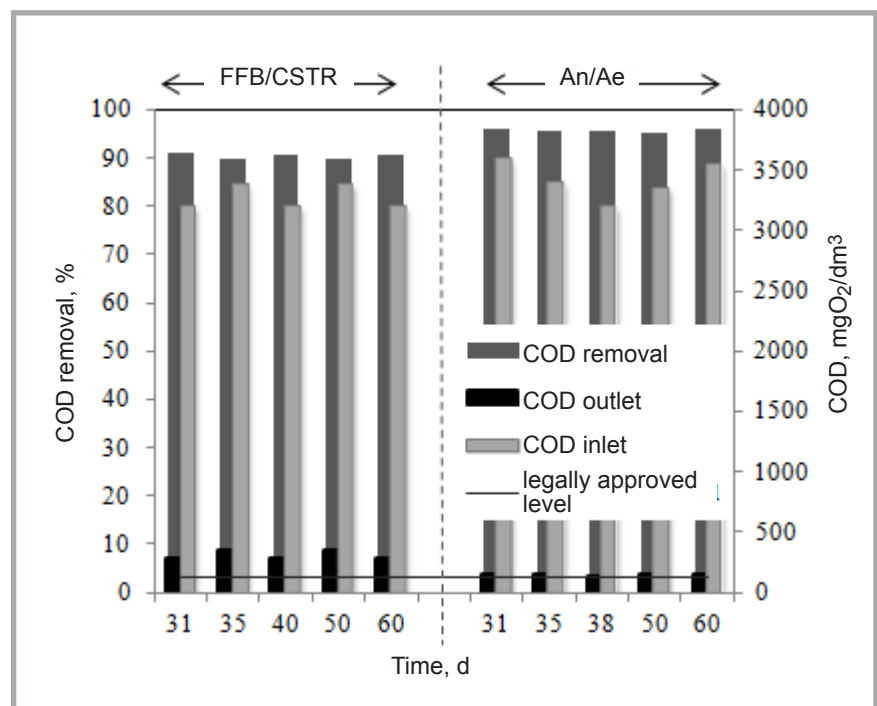


Figure 5. COD time changes in FFB/CSB and An/Ae.

water soluble azo dye by bacterial culture isolated from dye house effluent was 6 – 9.

Although both bioreactor systems examined had biogas collecting and volume measurement systems, the biogas production was very low and could not have been measured. On the basis of indirect biogas production assessment in

OxiTops, it can be noticed that biogas production from real textile wastewater was low and comparable with the control sample. The nanofiltration concentrate had a higher biogas yield than real textile wastewater, despite the fact that biogas production started after 50 h of the test, which can indicate the decomposition of non-easily biodegradable compounds (*Figure 6*).

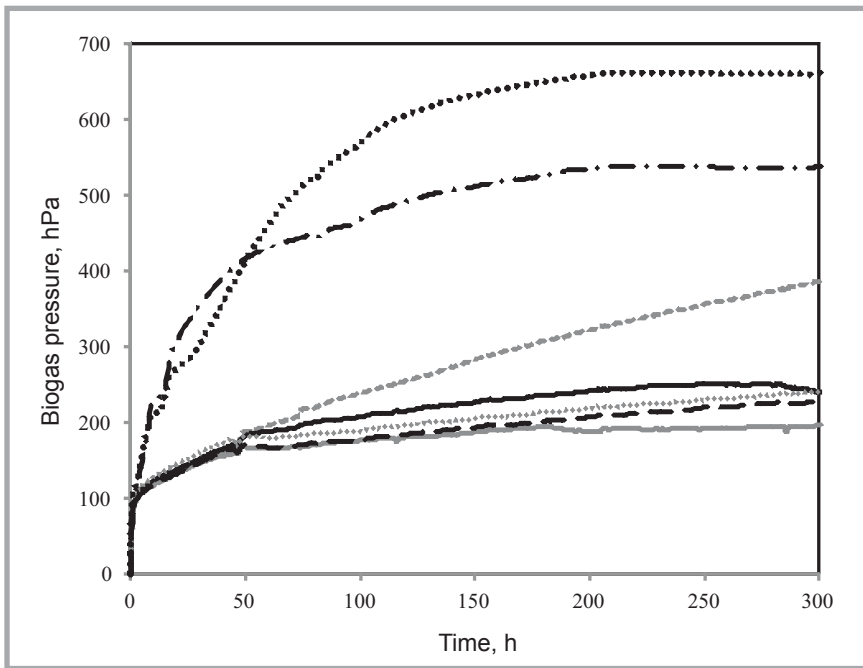


Figure 6. Indirect biogas measurement: from the top dotted black line - nanofiltration concentrate with synthetic wastewater; dash dot line - control with synthetic wastewater; dashed grey line – nanofiltration concentrate, solid black line – real textile wastewater I, dotted grey line – real textile wastewater II, dashed black line- real textile wastewater III, solid grey line - control.

The nanofiltration concentrate did not inhibit biogas production in the case of samples with an additional carbon source. Spagni et al. [23] reports the severe effect of the azo dye Reactive Orange 16 on biogas production, but the dye load applied was significantly higher (up to 3200 mg·dm⁻³) than in nanofiltration concentrate (120 mg·dm⁻³). A lack of biogas production in bioreactors can be explained by the high pH, which despite initial adjustment to neutral had a tendency to increase to a level higher than optimal for ADS. Furthermore during the processes in bioreactors, intermediate products or aromatic amine accumulate, which can increase the inhibitory effect along with the time of operating [24].

However, the effect of azo dyes on biogas production has been discussed by many authors [10, 22 - 24], and due to the varied structure of dyes one conclusion cannot be drawn. Some authors have observed the inhibition of biogas production [23], and others not [10]. Nevertheless most papers deal with synthetic textile wastewater or dye solution. The number of papers concerning the influence of real textile wastewater on methanogen activity is limited. Senthilkumar et al. [25] observed a sudden decrease in biogas production at a higher ratio of textile wastewater. Tapioca sago wastewater was also used as a co-substrate in a pilot scale two-phase Upflow Anaerobic Sludge Blanket (UASB) reactor.

SEM analysis of the activated carbon surface

In FFB, the activated carbon acted both as supporting material for microbial growth and as a regenerable redox mediator. SEM observation confirmed that the activated carbon provided good conditions for microbial growth (**Figure 7**). The activated carbon surface was covered with microbial extracellular polymeric substances (EPS) (**Figure 7.b**). According to Kokabian et al. [26], the production of EPS assessed on the basis of sludge free turbidity during the decolourisation of Reactive Black 5 increased with an increase in salt concentration, probably as a protective response of the mixed bacterial population. EPS makes up the intercellular space for microbials to aggregate and form the structure and architecture of the biofilm matrix. Biofilms maintain optimal pH conditions, localised solute concentrations and the redox potential, allowing cells to improve mineralisation processes [27]. On the other hand, results obtained by Rios-Del Toro et al. [28] suggests that the biofilm formed on activated carbon fibre (ACF) significantly reduced its redox-mediating capacity. Nevertheless due to biofilm formation, the FFB bioreactor was more efficient than the An bioreactor in terms of the volume of nanofiltration concentrate biodegraded per day of operation. In both anaerobic bioreactors examined the redox potential was below -350 mV. No significant changes in the redox potential along with biofilm formation were observed. A top layer of the biofilm formed on the activated carbon could have been easily removed at low shear; the second layer of biofilm can be seen in **Figure 7.c**. The same results were observed by Walter et al. [29] and Coufort [30], who tested different substrates for biofilm adhesion and with both aerobic and anaerobic bacteria.

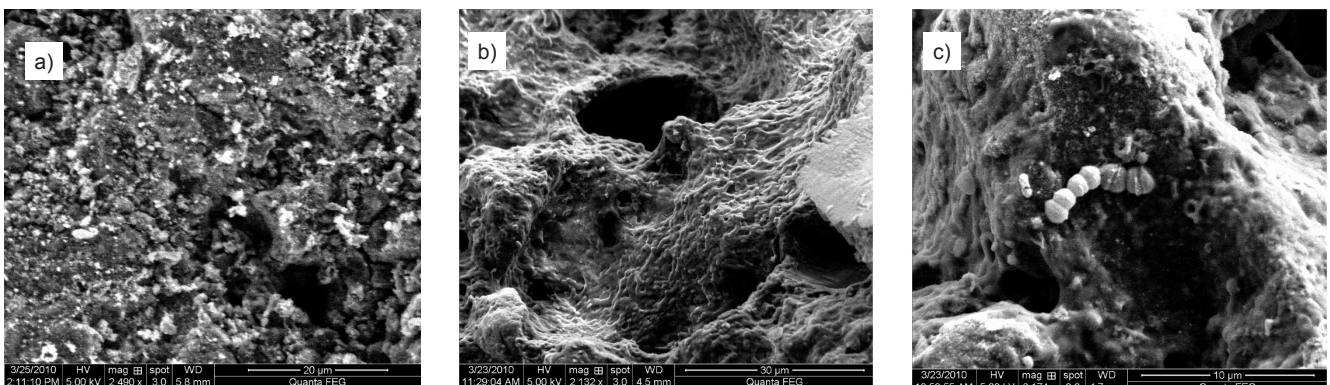


Figure 7. SEM microphotographs a) activated carbon, b) and c) with the biofilm and bacterial colony after removal of the first layer of biofilm.

They drew a conclusion that in the biofilm, three layers can be distinguished: a top layer - the most fragile and easily detached (60% of the initial biomass), an intermediate layer, and a third residual layer remained on the surface, which could only be detached at very high shear (20% of the initial biomass each). Such a structure is regarded as a general characteristic of biofilms [29, 30].

■ Conclusions

The implementation of membrane techniques for the reclamation and recycling of technological water in textile plants generates a concentrate which could be biodegraded. The results presented suggest the possibility of the application of anaerobic/aerobic systems for textile wastewater treatment: decolourisation and purification. The fixed film bioreactor reactor enables the implementation of continuous treatment of the nanofiltration concentrate as well as real textile wastewater. Aromatic amine released as a result of azo bond cleavage was effectively removed by the activated sludge in the aerobic reactor. Due to the significant variety of textile wastewater composition, anaerobic decolourisation cannot be treated as a stable biogas source. Further improvement of the bioreactors systems proposed and integration of nanofiltration and biological processes might allow the textile industry to meet the requirements of UE policies for environmentally sustainable development due to the closing of the water cycle and reduction of wastewater potential toxicity.

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