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Combination of adsorption and biological treatment in a SBR for colour elimination in municipal wastewater with discharges of textile effluents

E. Ferrer-Polonio*, A. Iborra-Clar, J.A. Mendoza-Roca, M.I. Iborra-Clar

Instituto de Seguridad Industrial, Radiofísica y Medioambiental, Universitat Politècnica de València, Camino de Vera s/n 46022, Valencia, Spain. Tel. +34 96 3877630; Fax: +34 96 3877639; emails: evferpo@posgrado.upv.es, evaferrerpol@gmail.com (E. Ferrer-Polonio), aiborra@iqn.upv.es (A. Iborra-Clar), jamendoz@iqn.upv.es (J.A. Mendoza-Roca), miborra@iqn.upv.es (M.I. Iborra-Clar)

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ABSTRACT

Discharge of textile wastewaters (WW) to municipal wastewater treatment plants (MWWTPs) entails the presence of colour in the final effluent. It causes a negative impact on the environment and, additionally, hinders an efficient disinfection by UV lamps. In this work, a combined process consisting of the addition of powdered activated carbon (PAC) to a sequencing batch reactor was studied. The main objective was to reduce WW colour in order to obtain transmittance values in the final effluent above 60%, measured at a wavelength of 254 nm, with the aim of ensuring disinfection with UV lamps. Experiments were performed with both simulated wastewater (SWW) including the azo dye Reactive Black 5 and WW from a MWWTP receiving discharges from textile mills. Biosorption increased the transmittance of the effluent around 25% for SWW and 24% for WW, in comparison with the values measured in the influent. The PAC concentrations for the achievement of a value of 60% in the transmittance of the treated water were 250 and 400 mg/L for the simulated effluent and for the WW, respectively. PAC had to be periodically added in order to cover its loss in the waste sludge.

Keywords: Colour removal; Powder activated carbon; Remazol Black 5; SBR

1. Introduction

Coloured effluents are produced by industries such as textile, leather and paper mills that use dyes in their production process. The presence of small amounts of dyes in water is highly visible and undesirable. Reactive dyes are the most common dyes used in textile industry, due to their advantages such as bright colours, excellent colourfastness and ease of application. Discharge of coloured wastewater (WW) from textile industry into municipal wastewater treatment plants (MWWTPs) with conventionally activated sludge processes entails coloured treated effluents. This is due to the fact that reactor biomass is not able to degrade reactive dyes [1]. Reactive dyes are normally azo-based chromophores combined with different types of reactive groups (e.g. vinyl sulphone, chlorotriazine, etc.) [2]. An operational problem is associated with the residual colour regardless of the eventual dye toxicity: the decrease in the efficiency of disinfection treatments by means of UV radiation. This compels the plant manager to take measures leading to colour removal before the disinfection stage.

*Corresponding author.

Upgrading MWWTPs for colour elimination is not an easy task. Many techniques reported in the bibliography (advanced oxidation techniques by Fenton's reagent [3] and ozone [4]) are effective. However, at industrial scale, the cost of these processes is very high. Besides, eventual formation of partially oxidized substances that could be dangerous has limited their application. Membranes [5,6] are also used in the processes that separate dyes from WW. Nevertheless, membrane fouling phenomena and reject streams management are drawbacks to take into account [7].

Anaerobic treatment is also successful for azo dyes removal. Nevertheless, breakdown of azo dye bonds results in the formation of aromatic amines, which can be more toxic than the original dye molecules [8,9]. Some authors have studied colour elimination by implementing a double biological stage (anaerobic/aerobic): an anaerobic process that degrades azo bonds to aromatic amine intermediates and an aerobic process for their total degradation [10–12]. In MWWTPs that are working with biological aerobic treatment, the implementation of an anaerobic reactor would require additional reactor volume and its efficiency would highly depend on temperature (optimal temperature of 36˚C).

The solution proposed in this paper is the addition of activated carbon in the aerobic biological reactor for colour removal. With this operating method, adsorption would form a part of the secondary stage, and not as a tertiary treatment. Activated carbon (AC) properties for colour adsorption have been reported by many studies [13,14].

Only a few studies address the application of powder activated carbon (PAC) in a bioreactor. These works mainly aim at the removal of micropollutants [15,16], hexavalent chromium [17] and the improvement in the removal of organic matter in high-polluted WWs like leachates [18]. Besides, it has to be highlighted that there are references focusing on colour removal in simulated wastewater (SWW) consisting of solutions of one or more dyes [19,20], but there are very few references that deal with real WW.

The novelty of this study is the application of PAC for colour elimination in a biological reactor, working with both SWW containing azo dye (Reactive Black 5) and WW collected from a MWWTP receiving textile industry effluents.

The experimental work consisted of three steps. The first one was a series of jar-tests with SWW to compare two commercial PACs to find out the concentration for achieving a transmittance of 60% at 254 nm (T_{254}). The second and the third steps corresponded to the experiments in the sequencing batch reactor (SBR) with PAC addition, treating SWW and MWWTP effluents, respectively.

2. Experimental

2.1. Chemicals

The chemicals used for preparing the SWW were peptone, meat extract (Panreac) and the dye, Reactive Black 5 (C.I. 55% dye, Sigma-Aldrich, RB5). RB5 was added to the reactor from a previously prepared 10% solution. Fig. 1 shows the chemical structure of the RB5.

Clarimex 061 (CHIEMIVALL) is a carbon made from pine wood, chemically activated with phosphoric acid. GMI 835/05 from GALAQUIM is a mineral carbon provided in a granular form. For the tests, the granular carbon was ground to a size similar to the Clarimex 061 (minimum of 80% lower than the 400 mesh size in US standard sieve, equivalent to 37 μm).

2.2. Activated sludge for the start-up

The activated sludge was taken from the bioreactor of a MWWTP, located in Valencia Region receiving discharges from a textile mills area. Thus, microorganisms adapted to the presence of colour. The initial concentration of the mixed liquor suspended solids (MLSS) in SBR was 1,500 mg/L.

2.3. SWW

SWW consisted of 270 mg/L of peptone and 270 mg/L of meat extract dissolved in tap water. Thus, an effective source of nitrogen and carbon was ensured. Its COD was 600 mg/L, similar to the value of the WW influent to the MWWTP. Ten milligram per litre of RB5 was added in order to mimic the transmittance at 254 nm of the influent to the MWWTP. Table 1 presents the SWW characterization.

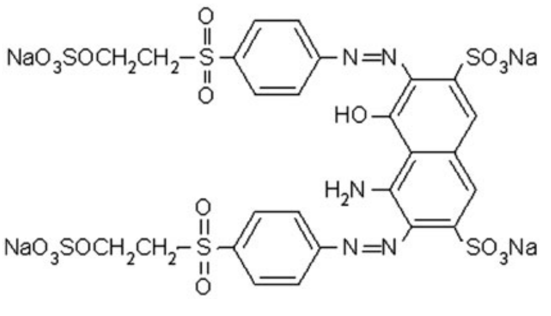

Fig. 1. Chemical structure Reactive Black 5.

Table 1
Wastewater characterization

Parameter	SWW	WW
COD (mg/L)	575–625	600–670
Conductivity (μS/cm)	1,330–1,550	1,860–2,280
pH	8–8.2	7–8
Temperature (°C)	20–25	20–25
Suspended solids (mg/L)	≈0	70–650
Transmittance 254 nm (%)	20.0–21.6	1.0–31.1

2.4. WW

WW was taken at the input of the biological reactor of the MWWTP. Its characterization is also presented in Table 1.

2.5. Analytical methods

Conductivity and pH were measured by means of analytical equipment from Crison. COD was determined by means of cell tests from Merck. MLSS were measured following APHA [21]. Transmittance was analysed with a model 8453 Hewlett-Packard spectrophotometer. pH, conductivity and MLSS were performed daily, meanwhile COD was measured twice a week.

2.6. Laboratory plant

A 25-L volume reactor was used as SBR for the experiments. The reaction volume used was 12 L in all the experiments. The SBR operation was controlled by timers programmed for changing the cycle at the required time. The mixing mechanism was provided via a Heidolph mechanical stirrer. Air was supplied by means of an air blower (Eheim de GmbH & Co. KG) for a maximum air-flow rate of 400 L/h and air was diffused through porous ceramic diffusers into the reactor. Dissolved oxygen was measured with a CRISON OXI 330 and its concentration was maintained between 2 and 3 mg/L in the aerobic phases. Masterflex pumps (MILLIPORE) were used for feeding the WW and extracting the effluent in the bioreactor. The plant scheme is shown in Fig. 2.

The laboratory SBR worked with three daily cycles of operation according to the strategy detailed in Table 2. As it can be seen, the hydraulic retention time was 1 d, i.e. the reactor was operated as an extended aerated activated sludge process maintaining a sludge retention time around 20 d. The volumetric exchange ratio (fill volume divided by reaction volume, VER) was 0.33. The reaction phase was divided into anoxic and aerobic phases.

3. Results and discussion

3.1. Work strategy

In the first part of the work, the commercial PAC was selected from two available carbons. For this, a series of jar tests with SWW was performed, testing different concentrations of AC (50–400 mg/L). Reaction time was 30 min. The pH and temperature values were the same as in the SBR operation. The reduction percentage of RB5 was calculated as the difference between the absorbance at the start and at the end of the test, measured at 599 nm, which is the maximum absorbance wavelength for the RB5 dye [14].

The operation strategy of the SBR was the same, regardless of the feeding WW. In both the cases (with SWW and with WW), there was a period of adaptation of the activated sludge to WW. The acclimation was completed once the stability of the effluent in pH, conductivity and COD was achieved and, consequently, biomass concentration increased in the reactor due to anabolic reactions. The next step was to determine the equilibrium T_{254} ($T_{254(eq)}$) in the SBR effluent after colour adsorption on the activated sludge flocs [22,23]. This step was carried out without PAC. Finally, the experiments with PAC addition were carried out and the first tested concentration was the optimum one calculated from previous jar tests.

As explained in Section 1, the aim of colour removal in this work was also based on solving the low efficiency of a subsequent disinfection with UV lamps. The loss of transmittance of the effluent to be disinfected reduces the radiation absorbed by the micro-organisms and, consequently, the disinfection efficiency. UV lamps work at 254 nm of wavelength since this is the optimum wavelength to damage the nucleic acids of the micro-organisms. It has to be highlighted that UV lamps suppliers recommend a $T_{254} \geq$ 60% for a high efficiency in the disinfection process. This is the reason why T_{254} has been selected as the control parameter.

3.2. PAC selection

Fig. 3 shows the results of the jar-tests. It is evident that GMI carbon was not appropriate for the elimination of RB5. This behaviour is in agreement with the AC characterization by the methylene blue adsorption test, since this index was 19 and 25 g/100 g for GMI and CLARIMEX 061 CAE, respectively. Ninety percent of dye concentration removal was achieved with a concentration of 300 mg/L of CLARIMEX CAE 061. Thus, it was decided to select CLARIMEX CAE 061 concentration as the initial one in the SBR experiments.

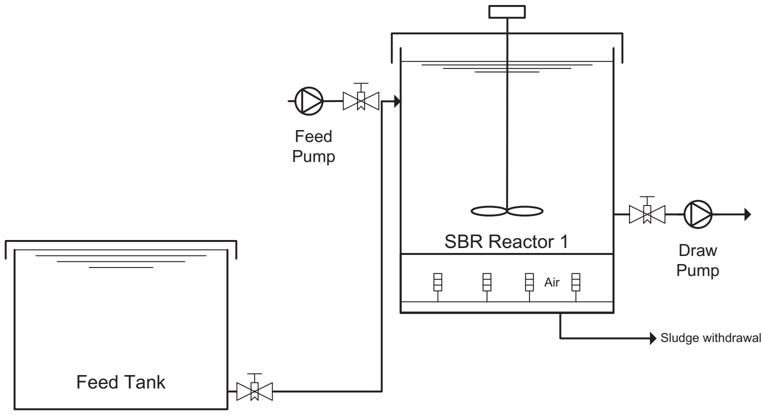

Fig. 2. Scheme of the SBR laboratory plant.

Table 2
SBR operation strategy of each cycle

	Time (min)
Filling	21
Anoxic reaction	60
Aerobic reaction	294
Sedimentation	90
Draw	7
Idle	8

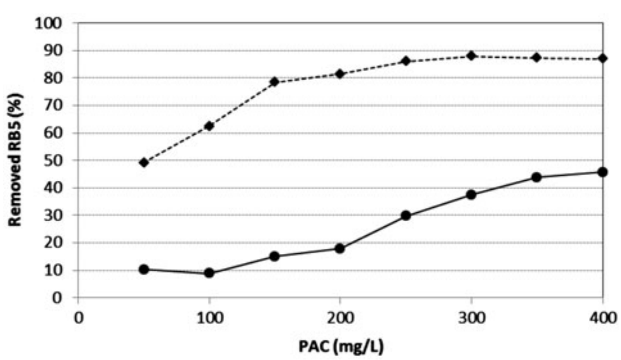

Fig. 3. RB5 removal by GMI/835-05 (•) and CLARIMEX 061 CAE (♦).

Galán et al. [1] reported that the Freundlich isotherm is better than the Langmuir isotherm for describing RB5 adsorption. The same behaviour was observed for the tested ACs. This suggests that the surface is not homogeneous and there is non-ideal adsorption. The results obtained with CLARMIEX CAE 061 fitted the Freundlich isotherm, whose equation is shown below.

$$q_e = K_F \cdot C_e^{1/n} \tag{1}$$

where q_e is the quotient between the masses of adsorbate and AC (mg/mg), C_e is the concentration of RB5 remaining in the solution (equilibrium concentration in mg/L) and K_F and n are the Freundlich constants. K_F is an indicator of the adsorption capacity and n is related to the intensity of the adsorption. Equation can be linearized by taking logarithms to find out the parameters K_F and $1/n$ (Fig. 4).

Thus, it was found $K_F = 0.026$ (mg RB5/mg PAC)·(mg PAC/L)$^{-0.7677}$ and $n = 1.30$. This value of $n > 1$ indicates favourable conditions for adsorption [24,25].

3.3. Experiments with SWW

In these experiments, sludge acclimation time was 7 d. Fig. 5 shows the effluent COD and MLSS

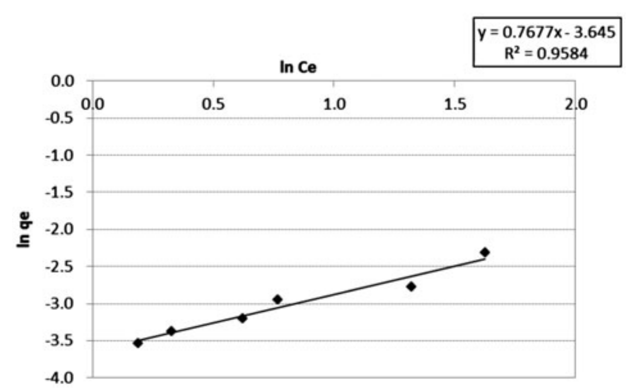

Fig. 4. Linearization of Freundlich equation for the data obtained with CLARIMEX 061 CAE.

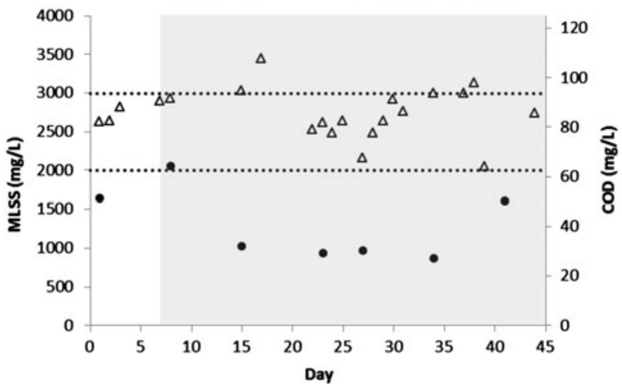

Fig. 5. Experiments with SWW; COD (•) and MLSS (Δ) evolution.

evolution in the 44 d of the experiment. The grey area defines the PAC testing period. The two horizontal lines delimit the optimum operating range considered for MLSS. There were three sludge withdrawals according to the aforementioned concentration range. After the sludge withdrawal, PAC was added to keep its concentration in the SBR.

The equilibrium transmittance was calculated for the first 7 d, once the biological process was considered to be stabilized without PAC addition. COD of the SBR effluent was always below 65 mg/L, which indicates a high efficiency of the biological process for organic matter elimination. Fig. 6 shows that T_{254} was practically constant during the first 7 d with an average value of 47.3%. This value was considered as $T_{254(eq)}$. As the T_{254} of the SBR influent (SWW) was around 21.6%, it can be stated that the activated sludge could absorb a certain amount of RB5 concentration since transmittance increased by 25.7% after the SBR treatment.

Fig. 6 shows the effluent transmittance evolution in the whole experiment. The horizontal line corresponds to the optimal transmittance to ensure the UV disinfection (60%). The three vertical lines show PAC additions. On day 8, PAC was added to the concentration of 300 mg/L in the SBR. As it can be seen in Fig. 5, COD decreased below 25–30 mg/L and in Fig. 6, transmittance of the SBR effluent increased sharply, from 45.7 to 70.5%. From that d on, T_{254} decreased gradually due to PAC losses in the waste sludge. Also, we detected small amounts of PAC in the effluent. The following PAC addition was carried out once the $T_{254(eq)}$ was reached again (after 15 operating days). Then, a new PAC dosing was performed in order to ensure the results reproducibility. After the new PAC addition, transmittance increased up to 65.3%, which means an increase of 24.2% in the transmittance value, very similar to that reached after the first PAC addition. In this operation period, transmittance decreased more lightly in comparison with the period after the first PAC addition. This was due to the increase in the PAC separation efficiency by settling, thereby no PAC concentration was observed in the effluent. After 16 d, the transmittance decreased to 48.1%.

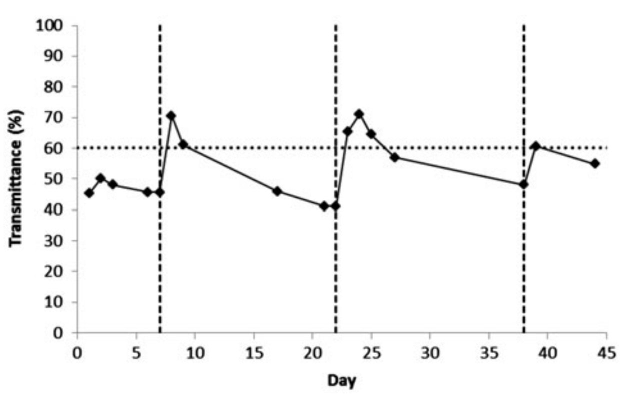

Fig. 6. T_{254} evolution in effluent SWW.

The third PAC addition was lower than the earlier one in order to optimize the PAC addition. By dosing 250 mg PAC/L, 60% of the required transmittance was achieved in the effluent required for the correct working of the UV lamps. Considering the PAC addition and the dye concentration in the SWW, it can be stated that by maintaining 250 mg/L of PAC (which implies to add the PAC lost in the sludge withdrawal), transmittance will remain around 60%.

3.4. Experiments with WW from the MWWTP

Sludge acclimation to the WW from the MWWTP was progressive in order to prevent eventual cellular degradation. In this way, at the beginning, WW was mixed with the SWW, without RB5, at a ratio of 1:1 (WW/SWW). Then, the mixture ratio was increased up to 3:1 and finally the SBR was fed only with WW. In Fig. 7, the vertical lines delimit the time intervals for each type of SBR feeding WW. Fig. 7 also shows the values of COD of the treated water and the MLSS in SBR. The two horizontal lines delimit the chosen operating range for MLSS and the grey area defines the PAC testing period.

Although the acclimatization was performed gradually, biomass was affected by the WW, which implied a decrease in the MLSS concentration and an increase in the soluble COD in the treated water. This was due to cell lysis phenomenon. Thus, MLSS concentration varied from 2,600 to 1,600 mg/L.

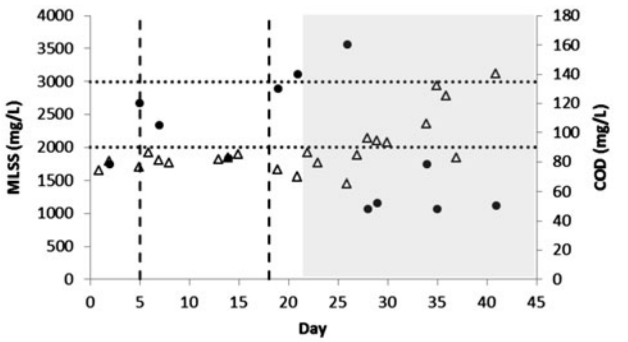

Fig. 7. Experiments with WW; COD (•) and MLSS (Δ) evolution.

The MLSS concentration reached values below the desired range until the 28th d. The COD concentration in the treated water exceeded the maximum concentration allowed by legislation (125 mg/L), when WW was fed exclusively to the SBR. Although the sludge acclimation to WW had not been reached, it was decided to start adding PAC in order to decrease the pollution and to improve the conditions for the survival of micro-organisms. PAC addition led to a considerable improvement of the process performance. COD values in the final effluent were around 50 mg/L and MLSS concentration increased as expected.

Fig. 8 shows the transmittance evolution of the treated WW with the operating day. The horizontal line means the optimal transmittance to ensure the UV disinfection (60%). The three vertical lines show PAC additions. The equilibrium transmittance was achieved from the 6th d and it was maintained until PAC addition. The measured $T_{254(eq)}$ was 28.3%. As the influent T_{254} was around 4.1%, it can be stated that the activated sludge process increased the transmittance in 24.2% due to adsorption on the flocs.

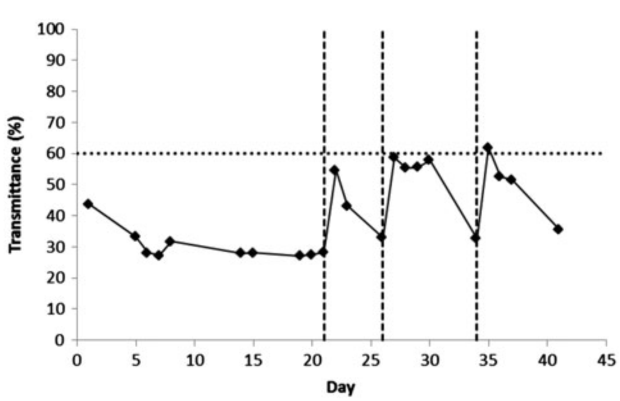

Fig. 8. T_{254} evolution in effluent from SBR fed with WW from MWWTP.

The time period with PAC addition was 21 d. Although the PAC optimum amount determined for SWW was 250 mg/L, a higher concentration was first added to the reactor due to the lower $T_{254(eq)}$, considering the same aimed transmittance of 60% in effluent. With 300 mg PAC/L on the first day after PAC addition, the reached transmittance was 54.6%. The new PAC addition was carried out with 400 mg/L on the 26th d that led to a transmittance of 59% in the final effluent. Then, a new PAC dosing of 400 mg/L was performed, in order to ensure the results reproducibility, on the 34th d. For this test, a transmittance of 62% was achieved.

4. Conclusions

This research reports an effective treatment for the removal of azo dyes in WW, combining biological oxidation with SBR and adsorption onto PAC. This is a possible solution for upgrading MWWTPs receiving discharges from textile mills without performing a tertiary treatment.

The adsorptive capacity of the sludge, reported in many papers, was verified. The transmittance of the SBR effluent, in both the experiments with SWW and WW remained constant once the equilibrium was reached. With SWW, the $T_{254(eq)}$ was around 46% and in WW, it was around 28%. In both the cases, the increase in the effluent transmittance with respect to the influent was very similar, around 25% for SWW and 24% for WW.

By adding a concentration of 250 mg/L PAC in the SBR, a transmittance of 60% in the final effluent was achieved when the SBR was fed with SWW. When the SBR was fed with WW from the WWTP, the added PAC had to be increased up to 400 mg/L to obtain the transmittance of 60%, required for further disinfection by UV lamps.

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Abbreviations

MWWTP	—	Municipal wastewater treatment plants
PAC	—	Powdered activated carbon
SBR	—	Sequencing batch reactor
RB5	—	Reactive Black 5
T_{254}	—	Transmittance of 60% at 254 nm
$T_{254(eq)}$	—	Equilibrium transmittance of 60% at 254 nm
MLSS	—	Mixed liquor suspended solids
COD	—	Chemical oxygen demand
SWW	—	Simulated wastewater
WW	—	Wastewater

References

[1] J. Galán, A. Rodríguez, J.M. Gómez, S.J. Allen, G.M. Walker, Reactive dye adsorption onto a novel mesoporous carbon, Chem. Eng. J. 219 (2013) 62–68.

[2] Z. Aksu, G. Dönmez, A comparative study on the biosorption characteristics of some yeasts for Remazol Blue reactive dye, Chemosphere 50 (2003) 1075–1083.

[3] C.C. Su, M. Pukdee-Asa, C. Ratanatamskul, M.C. Lu, Effect of operating parameters on decolorization and COD removal of three reactive dyes by Fenton's reagent using fluidized-bed reactor, Desalination 278 (2011) 211–218.

[4] P. Colindres, H. Yee-Madeira, E. Reguera, Removal of Reactive Black 5 from aqueous solution by ozone for water reuse in textile dyeing processes, Desalination 258 (2010) 154–158.

[5] R.M. Ribeiro, R. Bergamasco, M.L. Gimenes, Membranes synthesis study for colour removal of a textile effluent, Desalination 145 (2002) 61–63.

[6] M. Riera-Torres, C. Gutiérrez-Bouzán, M. Crespi, Combination of coagulation–flocculation and nanofiltration techniques for dye removal and water reuse in textile effluents, Desalination 252 (2010) 53–59.

[7] B. Van der Bruggen, G. Cornelis, C. Vandecasteele, I. Devreese, Fouling of nanofiltration and ultrafiltration membranes applied for wastewater regeneration in the textile industry, Desalination 175 (2005) 111–119.

[8] P. Kariyajjanavar, N. Jogttappa, Y.A. Nayaka, Studies on degradation of reactive textile dyes solution by electrochemical method, J. Hazard. Mater. 190 (2011) 952–961.

[9] A. Cardenas-Robles, E. Martinez, I. Rendon-Alcantar, C. Frontana, L. Gonzalez-Gutierrez, Development of an activated carbon-packed microbial bioelectrochemical system for azo dye degradation, Bioresour. Technol. 127 (2013) 37–43.

[10] F. Harrelkas, M.N. Pons, O. Zahraa, A. Yaacoubi, E.K. Lakhal, Application of a sequential system for textile dyes degradation: Comparison between azo and phtalocyanine dyes, Water Sci. Technol. 55 (2008) 107–114.

[11] R.M. Melgoza, A. Cruz, G. Buitrón, Anaerobic/aerobic treatment of colorants presents in textile effluents, Water Sci. Technol. 50 (2004) 149–155.

[12] S. Venkata Mohan, N. Rao, P.N. Sarma, Simulated acid azo dye (Acid black 210) wastewater treatment by periodic discontinuous batch mode operation under anoxic–aerobic–anoxic microenvironment conditions, Ecol. Eng. 31 (2007) 242–250.

[13] V. Gómez, M.S. Larrechi, M.P. Callao, Kinetic and adsorption study of acid dye removal using activated carbon, Chemosphere 69 (2007) 1151–1158.

[14] W.M. Alvin, P. Barford John, G. McKay, A comparative study on the kinetics and mechanisms of removal of Reactive Black 5 by adsorption onto activated carbons and bone char, Chem. Eng. J. 157 (2010) 434–442.

[15] X. Li, F.I. Hai, L.D. Nghiem, Simultaneous activated carbon adsorption within a membrane bioreactor for an enhanced micropollutant removal, Bioresour. Technol. 102 (2011) 5319–5324.

[16] D. Serrano, S. Suárez, J.M. Lema, F. Omil, Removal of persistent pharmaceutical micropollutants from sewage by addition of PAC in a sequential membrane bioreactor, Water Res. 45 (2011) 5323–5333.

[17] A.M. Orozco, E.M. Contreras, N.E. Zaritzky, Effects of combining biological treatment and activated carbon on hexavalent chromium reduction, Bioresour. Technol. 102 (2011) 2495–2502.

[18] F.J. Rivas, F.J. Beltran, O. Gimeno, J. Frades, F. Carvalho, Adsorption of landfill leachates onto activated carbon equilibrium and kinetics, J. Hazard. Mater. 131 (2006) 170–178.

[19] S. Sirianuntapiboon, O. Sadahiro, P. Salee, Some properties of a granular activated carbon-sequencing batch reactor (GAC-SBR) system for treatment of textile wastewater containing direct dyes, J. Environ. Manage. 85 (2007) 162–170.

[20] K.D. Belaid, S. Kacha, M. Kameche, Z. Derriche, Adsorption kinetics of some textile dyes onto granular activated carbon, J. Environ. Chem. Eng. (2013) 496–503.

[21] APHA, Standard Methods for the Examination of Water and Wastewater, 20th ed., American Public Health Association, Washington, DC, 1998.

[22] Z. Aksu, A.B. Akın, Comparison of Remazol Black B biosorptive properties of live and treated activated sludge, Chem. Eng. J. 165 (2010) 184–193.

[23] R.G. Saratale, G.D. Saratale, J.S. Chang, S.P. Govindwar, Bacterial decolorization and degradation of azo dyes: A review, J. Taiwan Inst. Chem. Eng. 42 (2011) 138–157.

[24] G. McKay, H.S. Blair, J.R. Gardner, Adsorption of dyes on chitin. I: Equilibrium studies, J. Appl. Polym. Sci. 27 (1982) 3043–3057.

[25] T.K. Saha, N.C. Bhoumik, S. Karmaker, M.G. Ahmed, H. Ichikawa, Y. Fukumori, Adsorption characteristics of Reactive Black 5 from aqueous solution onto chitosan, CLEAN—Soil, Air, Water 39 (2011) 984–993.