Advanced oxidation for aromatic amine mineralization after aerobic granular sludge treatment of an azo dye containing wastewater

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Abstract
Advanced oxidation processes (AOP), namely ozonation and UV irradiation, were tested to evaluate the possibility of complete mineralization of the metabolites resulting from biodecolorization of the azo dye Acid Red 14. These were present in the effluent collected from an aerobic granular sludge bioreactor operated on a 6-h cycle, including a 5-h reaction step comprising 2 stages, a 2-h anaerobic followed by a 3-h aerobic. This effluent was examined by high performance liquid chromatography (HPLC) and 4-amino-1-naphthalenesulfonic acid (4A1NS) was found to be the major metabolite, derived in a stoichiometric amount from the complete bioreduction of the added dye. AOP application results indicated that 85% removal of the 4A1NS amine can be obtained after 5 minutes of ozonation or 20 minutes of UV irradiation. UV-visible spectral and HPLC profiles were different in the effluents from the two AOP, suggesting different conversion patterns, namely concerning aromatic compounds. COD removal was found to be negligible, under 5%, in both cases. However, a simulated recirculation of the AOP-treated effluents back to the biological treatment stage resulted in significant COD removal yields in the 15-20% range. It was concluded that the tested, simple AOP are promising post biological treatment options for promoting the mineralization of the tested azo dye's persistent metabolites.

Keywords
Advanced oxidation processes; ozonation; UV irradiation; aromatic amines; azo dye; aerobic granular sludge

INTRODUCTION

The use of biological processes combining anaerobic and aerobic steps for the treatment of textile effluents containing azo dyes has been successfully demonstrated, achieving the removal of both color and organic load. Color is mostly removed in the anaerobic phase due to azo dye bioreduction and the aerated phase is generally responsible for most of the organic load removal. Lately, aerobic granular sludge (AGS) systems have also been tested for this purpose (Muda et al. 2010; Kolekar et al. 2012; Mata et al. 2015). AGS systems can result in cost savings when compared to flocculent activated sludge processes, due to the high biomass concentrations that can be attained, resulting in shorter cycle times and reduced reactor volumes. However, the degradation of recalcitrant amines resulting from the bioreduction of the azo dyes is often not achieved in activated sludge systems, leading to the need to evaluate combined biological and post-treatment solutions for this purpose (Jonstrup et al., 2011). In fact, despite advanced oxidation processes (AOP) being traditionally used as pre-treatments in industrial wastewater remediation (Oller et al. 2011), some authors have recently proposed combined systems for textile wastewater treatment in which the first stage is biological and the AOP is applied as post-treatment (Garcia-Montano et al. 2008; Kim et al. 2011; Lotito et al. 2012).

In this context, advanced oxidation processes (AOP) involve the use of an oxidizing agent such as ozone (O_3) or hydrogen peroxide (H_2O_2) to convert the dyes or their metabolites to less harmful products. Among AOP agents, ozone is the most widely used because of its higher reactivity (e.g.,

its standard oxidation-reduction potential is 2.07 V, while that of H_2O_2 is 1.78 V) and has been shown to efficiently remove color even at low dosages (Robinson et al. 2001; Alaton et al. 2002; dos Santos et al. 2007; Solís et al. 2012). Ozone may react directly with the dye molecules, in addition to its action via the hydroxyl radical HO• formed from O_3 reactions in aqueous media (2.8 V standard oxidation-reduction potential; Solis et al. 2012). UV radiation based methods, either alone or aided by the presence of various oxidants, such as H_2O_2, or catalysts, such as TiO_2, have also shown a high efficiency in dye color removal. The use of solar radiation, instead of mercury lamps, as a source of UV light for the treatment of colored effluents has also attracted the attention of researchers (dos Santos et al. 2007).

The objective of the present study was to assess the possibility of applying AOP, in simple configuration, as a post-treatment of a simulated textile effluent, pre-treated in a sequential batch bioreactor system with aerobic granular sludge. The AOP process would not have the objective of removing color, previously removed with high efficiency in the anaerobic stage of the bioreactor operating cycle, but of promoting the mineralization of the azo dye metabolites remaining in the effluent from the AGS bioreactor. In particular, the conversion of an aromatic amine metabolite, found to be stable and recalcitrant to biodegradation in the AGS stage, was examined throughout the application of ozonation and UV irradiation post-treatments. The recirculation of the AOP-treated effluent back to the AGS bioreactor was also tested, in order to assess the possibility of improvement of the overall removal of organic load from the fed wastewater.

MATERIALS AND METHODS

AGS biologically treated wastewater

The AGS-treated wastewater sample was obtained by pooling together the biologically treated effluent produced in several cycles of an AGS sequencing batch reactor (AGS-SBR), collected along a period of 2 days. The latter was operated in 6-h cycles, including a 5-h reaction stage (2-h anaerobic followed by 3-h aerobic), and fed with a simulated textile wastewater containing the azo dye Acid Red 14 (AR14; Chromotrope FB, dye content circa 50%, Sigma-Aldrich, USA) at 40 mg/L, a starch-derived sizing agent (Emzise E1, Emsland-Stärke GmbH, Germany) at 1000 mg COD/L, and macro and micronutrients as described previously (Mata et al., 2015). During this sampling period, the AGS-SBR kept stable color and organic load removal yields, both above 80%, with a sludge age value of approximately 15 days. The collected effluent pool was filter clarified with qualitative filters (type 1-Qualitative, Whatman, UK) and stored at 4°C until use.

Ozonation post-treatment

For the ozonation experiments, a 60-watt Electronic Ozonizer, model HLO-820A (Hailea, China) was used, with a maximum gas flow rate of 15 L/min, corresponding to 2000 mgO_3/h. It was operated at 5 L/min and applied to an effluent sample volume of 800 mL. The reactor was a 1-L glass cylinder with magnetic stirring and the ozone-containing gas stream was fed at the bottom using an acrylic diffuser. Ozone was fed continuously for 60 minutes, with 15-mL samples collected at 0, 5, 10, 20, 30, 45 and 60 min.

UV irradiation post-treatment

For the UV irradiation experiments, a 150-watt, medium pressure mercury lamp (type TQ150, Heraeus Noblelight, Germany) was used, emitting polychromatic radiation in the 200-600 nm range. The lamp was mounted on the central axis of a 850-mL photoreactor, surrounded by a quartz jacket with cooling water circulation (Heraeus Noblelight, Germany). Such arrangement allowed the effluent sample temperature to be kept at about 25°C during the irradiation runs. A volume of 600 mL of effluent was irradiated continuously for up 30 minutes, under magnetic stirring, with 15-

mL samples collected at 0, 5, 10, 20 and 30 min.

Simulation of the recirculation of the AOP post-treated effluent to the AGS-SBR

After application of the AOP treatment, a part of this post-treated effluent was subjected to a single biological treatment cycle, consisting of a 2-h anaerobic reaction phase followed by a 3-h aerobic phase, simulating the process of recycling this effluent to the AGS-SBR reactor. The concept is represented in Figure 1. The tested effluents were collected after ozonation and UV irradiation treatments with durations of 60 and 30 min, respectively. The biological reactor used was a 1-L glass cylinder. The anaerobic phase was operated under magnetic stirring and the aerobic phase under magnetic stirring plus aeration, using an acrylic diffuser and a compressor (model AC-9602, Aquapor, Portugal).

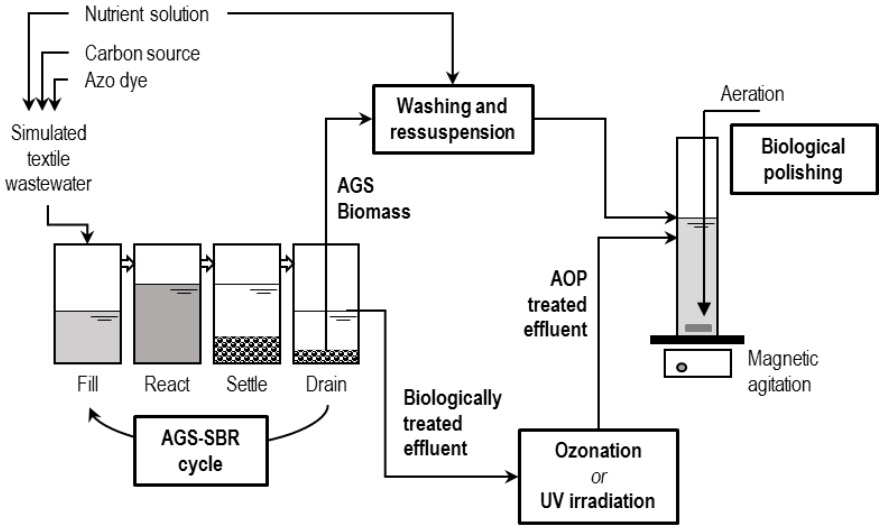

Figure 1. Scheme of the experimental steps for simulating the recirculation of the AOP-treated effluents to the AGS-SBR.

The AGS biomass was collected from the AGS-SBR reactor and washed with the solution of macro and micronutrients before being used in the simulated recirculation tests. A 100-mL portion of biomass suspension in nutrient solution (with no added carbon source) was added to a 250-mL sample of effluent from each of the AOP processes. The resulting biomass concentrations were 3.2 and 3.5 g TSS/L in the bioreactor tests with samples of the ozonation and UV irradiation effluents, respectively.

Analytical methods

The AOP and biological treatment experiments were followed by analysing samples from the reaction liquid using HPLC, spectral analysis in the UV-visible range (UV-vis), chemical oxygen demand (COD) and total suspended solids (TSS, only for bioreaction samples). For HPLC, UV-vis and COD analyses, the samples were previously clarified by centrifugation (10 min, 4000 rpm). Soluble COD and TSS were determined following standard procedures (APHA, 1998). The UV-vis spectra were acquired between 190 and 800 nm against distilled water in a Specord 200 spectrophotometer (Analytik Jena, Germany), with a quartz cell of 1 cm optical path length. HPLC analyses were performed using a Merck-Hitachi (Germany) system comprising an L-6200A Intelligent Pump, an L-4250 UV–vis Detector and an L-7200 Autosampler with a D7000 Interface for computer control, fitted with a reversed phase RP-18, LiChroCART 250-4 column (Merck, Germany), with spectrophotometric detection at 220 nm. The mobile phase, fed at 0.7 mL/min, was composed of sodium phosphate buffer (25 mM, pH 5.5) and acetonitrile, run on a 30-min linear

gradient from 100:0 to 50:50 (v/v), followed by a 5-min linear gradient up to 15:85 (v/v), and ending with a step back to 100:0, kept for a further 10-min period. Standard solutions of the dye AR14 and of the amine 4-amino-1-naphthalenesulfonic acid (4A1NS) (Sigma-Aldrich, USA) in distilled water were run for their respective peak identification and concentration calibration against peak area. The standard deviation (STD) of the method was determined with values obtained from 10 replicates. STD values were 5.7% for 1 mg/L AR14 and 2.7% for 1 mg/L 4A1N and 1.0% and 0.5%, respectively for their 10 mg/L standards. Each sample was measured in duplicate and mean values are presented. Residual ozone concentrations in the liquid medium were measured using a reagent kit, ref. AP 056 (range 0-2 mg O_3/L), and a Photometer model 5000, both from Palintest (UK).

RESULTS AND DISCUSSION

AGS-SBR biologically treated wastewater samples

During the sampling period for the AOP runs, the AGS-SBR maintained stable color and COD removal yields, giving a residual COD level around 170 mgO_2/L. Although color removal was not complete, as measured at 515 nm (wavelength of maximum absorbance for the dye AR14), HPLC results showed that the added dye was completely converted. The residual color was attributed to reactions involving the amine 2-amino-4-hydroxy-1-naphthalenesulfonic acid (2A4H1NS) resulting from azo bond reduction on AR14 (Figure 2), which was found to be unstable, possibly undergoing auto-oxidation (Mata et al. 2015). Also from the HPLC results, the stable metabolite 4-amino-1-naphthalenesulfonic acid (4A1NS) was identified and quantified in an approximately stoichiometric amount in relation to that of the reduced AR14 (Figure 2), indicating that no bioconversion of this amine had occurred in the aerated phase of the AGS-SBR cycle. Thus, this amine was subsequently used as an indicator of the performance of the AOP post-treatments in the conversion of dye metabolites.

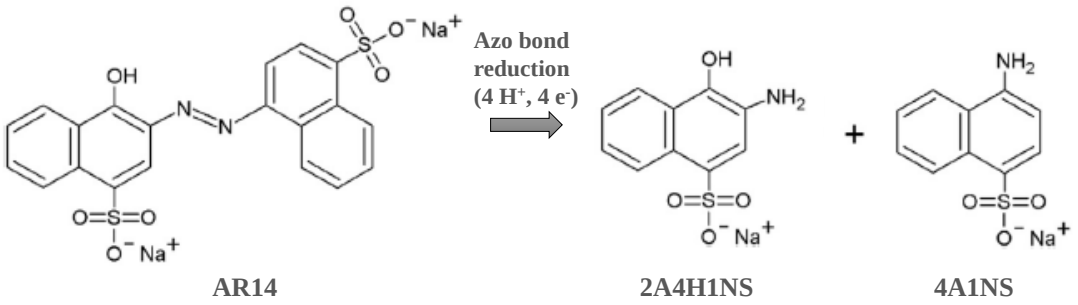

Figure 2. Chemical structures of the azo dye Acid Red 14 (AR14) and of the two aromatic amines formed during the azo bond reduction reaction, 2-amino-4-hydroxy-1-naphthalenesulfonic acid (2A4H1NS, *orto*, unstable amine) and 4-amino-1-naphthalenesulfonic acid (4A1NS, *para*, stable amine).

Ozonation post-treatment

The AGS-SBR effluent was subjected to a batch ozonation process with a continuous feed of ozone-enriched gas stream for a period of up to 60 minutes, at room temperature. Figure 3 shows the results obtained for the residual concentration of 4A1NS in the reaction medium (as determined by HPLC) along the operational time, and the corresponding UV-vis spectra (190-800 nm) of selected samples. The residual level of ozone measured in the liquid was 0.2 mg O_3/L at the end of experiment (60 min).

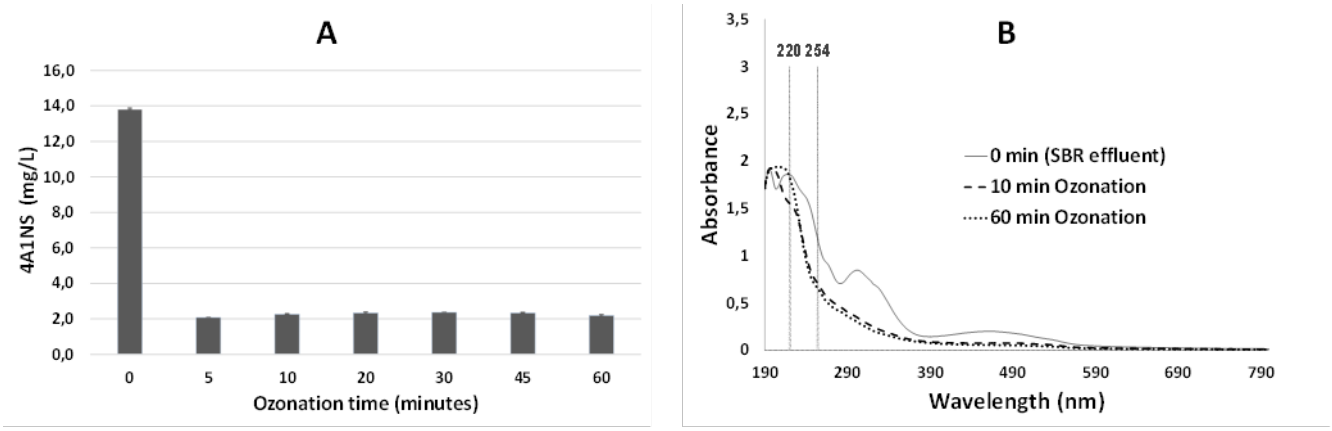

Figure 3. **A:** Evolution of the 4A1NS amine concentration as determined by HPLC along time during the ozone treatment applied to the AGS-SBR effluent. **B:** UV-vis spectra of samples taken at time 0 (original AGS-SBR effluent) and times 10 and 30 min during the ozonation experiment.

Already at 5 min of ozonation the conversion of the 4A1NS amine was apparently completed, down to a final level corresponding to 85% removal. Further conversion of the residual amine was not observed even with ozonation prolonged to 60 min (Figure 3A). The UV-vis spectra profiles clearly show the disappearance of the characteristic amine peak at 320 nm (Figure 3B), in accordance with the chromatographic results. It also appears that prolonging the ozonation from 10 to 60 minutes increased the absorbance in the 200-230 nm range, suggesting additional oxidative transformation of the residual organics.

Figure 4 shows HPLC chromatograms from the samples of Figure 3B. Observing these, in addition to the disappearance of major peaks, a general tendency for retention time reduction in the residual peaks is observed, suggesting conversions into lower molecular weight and more hydrophilic products. The peak at a retention time of 3.3 minutes (already detected at 20 minutes of ozonation, results not shown) seen in the 60-min chromatogram was not identified. It did not match a standard of 4-hydroxy-1-naphthalenesulfonic acid, a possible product of 4A1NS oxidation, since this appeared at a retention time of 19 min, in the HPLC analysis conditions.

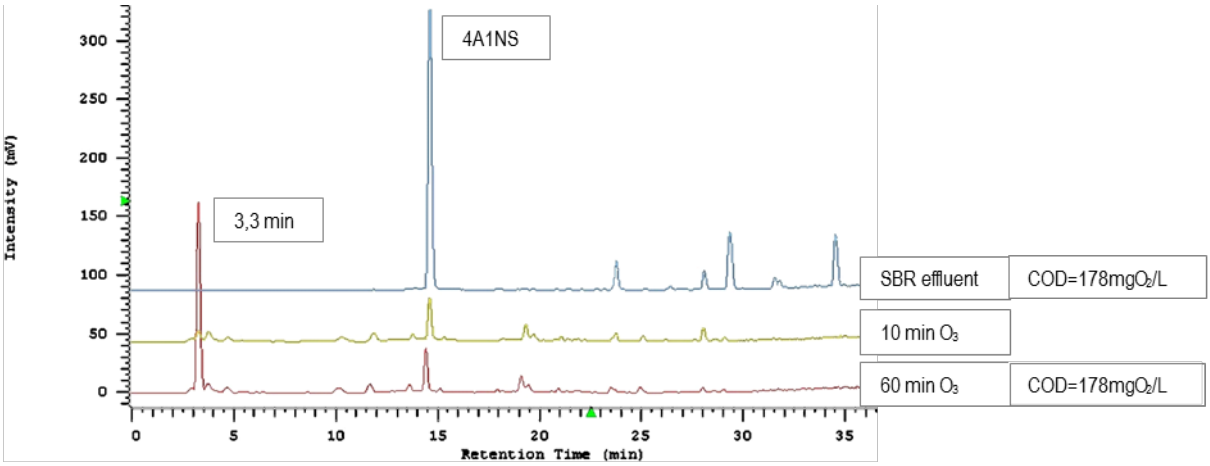

Figure 4. HPLC chromatograms of samples taken at different times during the ozonation treatment applied to the AGS-SBR effluent. COD values measured on the initial and final process samples are also presented.

As can be also seen in Figure 4, the COD value was unchanged after the 60-min ozonation period. It can thus be concluded that mineralization of the organic solutes present in the effluent was negligible.

In previous studies, the fate of recalcitrant amines was not followed in ozonation processes applied to effluents from bioreactors where azo dye decolorization had occurred (Baban et al. 2003; Garcia-Montano et al. 2008; Kim et al. 2011; Lotito et al. 2012). However, Baban et al. (2003) reported that 10 minutes of ozonation produced an effluent with no toxicity, possibly related to the conversion of intermediate amines. Likewise, Garcia-Montano et al. (2008) indicate that the products resulting from the anaerobic treatment of an effluent containing an azo dye were more toxic than the original components, but that after ozonation the effluent became non-toxic. These authors reported values of 48% and 83% for the mineralization yield of the anaerobic reactor effluent components in ozonation experiments run at the acid-neutral pH range and at a pH value of 10.5, respectively. The same authors also indicate that the difference was due to the oxidizing agent being the ozone molecule in the former case, and the hydroxyl radical (HO•) in the latter, with a higher oxidation-reduction potential than ozone (Solis et al., 2012; Robinson et al. 2001). In the present study, ozonation was carried out at neutral pH so the oxidizing agent was probably mostly ozone. Although toxicity testing has not been performed, 85% conversion of the recalcitrant aromatic amine 4A1NS was achieved, demonstrating the effectiveness of this post-treatment as a supplement to the biological treatment.

UV irradiation post-treatment

A sample of the AGS-SBR effluent pool was subjected to continuous UV irradiation for a period of up to 30 minutes. Figure 5 shows the results obtained from the HPLC measurement of 4A1NS concentration in samples taken along the irradiation time. UV-vis spectra (190-800 nm) of selected samples taken along this experiment are also shown.

From the results, a period of 20 min of irradiation is required to achieve a residual 4A1NS concentration of 2 mg/L (Figure 5A), corresponding to the 85% removal yield obtained in the ozonation experiment. UV-vis spectra of the samples confirm the previous result, since at 10 min of irradiation the 4A1NS characteristic peak at 320 nm is still identifiable.

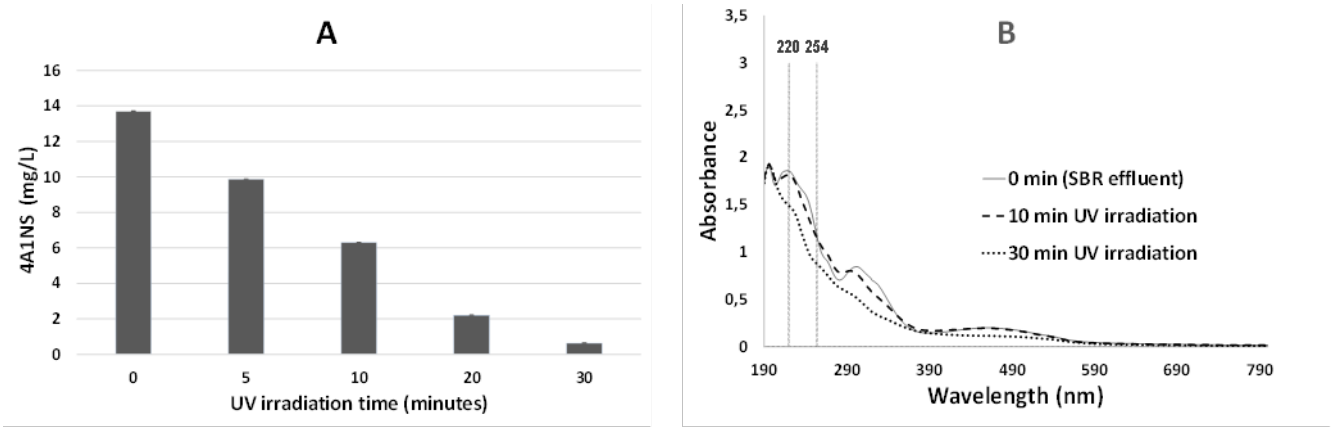

Figure 5. **A:** Evolution of the 4A1NS amine concentration as determined by HPLC along time during the UV irradiation treatment applied to the AGS-SBR effluent. **B:** UV-vis spectra of samples taken at time 0 (original AGS-SBR effluent) and times 10 and 30 min during the UV irradiation experiment.

A total period of 30 min of UV irradiation appears to further eliminate, in addition to the examined

amine (Figure 5A), almost all the metabolites detected in the chromatogram from the original effluent, as shown in Figure 6. In this case, in contrast to what happened in the ozonation experiment, no new oxidation products appear in significant amounts in the chromatograms from the irradiated samples. The UV-vis spectra profiles (Figure 5B) also show a decrease in absorbance values in the 220-254 nm range, often attributed to aromatic compounds.

COD reduction (Figure 6) through the application of UV irradiation was very low (corresponding to less than 5% elimination), showing, as observed for the ozonation process, that mineralization was negligible.

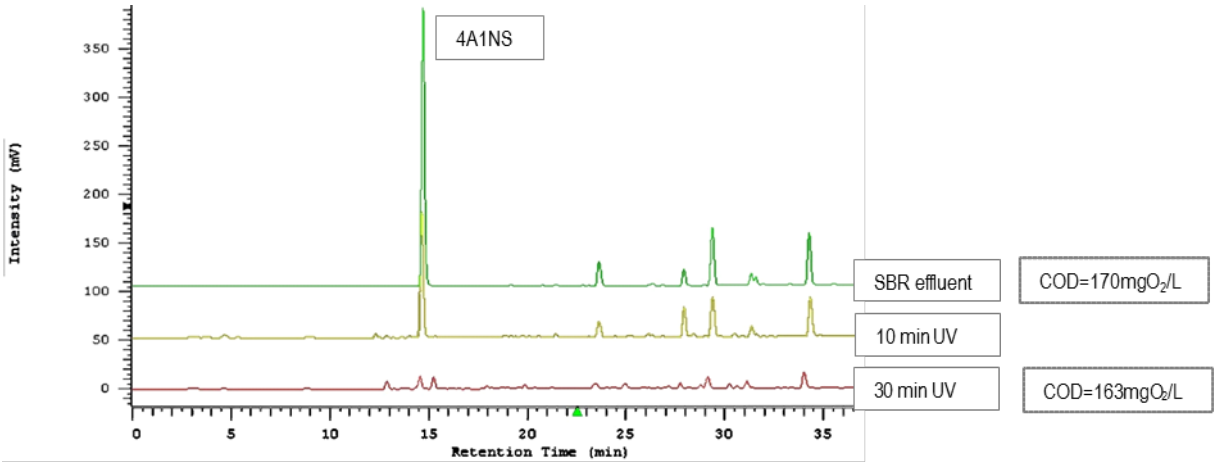

Figure 6. HPLC chromatograms of samples taken at different times during the UV irradiation treatment applied to the AGS-SBR effluent. COD values measured on the initial and final process samples are also presented.

Similar studies, namely, UV irradiation as a post-treatment for biologically treated textile effluents containing azo dyes, were not found in the literature. In one reported study (Dokuzoglu and Alkan 2010), UV irradiation was used as a pre-treatment before biological treatment, together with H_2O_2 in order to generate hydroxyl radicals, achieving an increase in dye biodegradability. The addition of H_2O_2 in the post-treatment by UV irradiation would have to be examined taking into consideration the additional cost and the possibly different profile of obtained products. UV irradiation alone is effective in removing the recalcitrant amine 4A1NS but an 85% removal yield requires a 20-min treatment, which can render it too costly due to the energy consumption.

Simulation of the recirculation of AOP post-treated effluents to the AGS-SBR

The AOP post-treatment options here examined were found to significantly alter the SBR treated effluent composition, namely by further converting at least one of the primary metabolites from azo dye bioreduction. However, the degree of mineralization attained, as measured by COD removal, was insignificant. Nevertheless, AOP treatment has been shown to improve the quality of textile effluents, biologically pre-treated or not, e.g., through reducing their toxicity (Garcia-Montano et al. 2008) or increasing their biodegradability (Dokuzoglu and Alkan 2010). The latter effects bring about the possibility of using the primary AGS-SBR to complete the treatment, by recirculating the AOP-treated effluent back into it. The sequential-batch operation provides flexibility as to the cycle time when the addition of the post-treated effluent would be most convenient. The test carried out in the present study aimed to simulate a situation in which the AGS-SBR cycle would be operated as usual, including the two reaction phases, except that it would be fed with the AOP-treated effluent instead of the raw textile wastewater. To further exclude effects from the residual organic load left

over from the AGS-SBR cycle, the granular biomass was collected, washed with nutrient solution free from carbon source and incubated with the AOP-treated effluent in a volumetric proportion calculated to reproduce a typical biomass concentration in the AGS-SBR.

The performance of AGS-SBR treatment of the effluent post-treated using each of the two types of AOP described above was followed by HPLC, COD quantification and UV-Vis spectrophotometry. Results from the latter are shown Figure 7. The UV-vis spectral profiles show no change from the biological treatment of the ozonised effluent, but for the UV-irradiated effluent the spectra exhibit a slight absorbance decrease in the 220-254 nm range, possibly indicating biological degradation of structures containing aromatic rings. A small COD removal effect was measured for both the pre-ozonised and the pre-irradiated effluents, 20 and 15%, respectively.

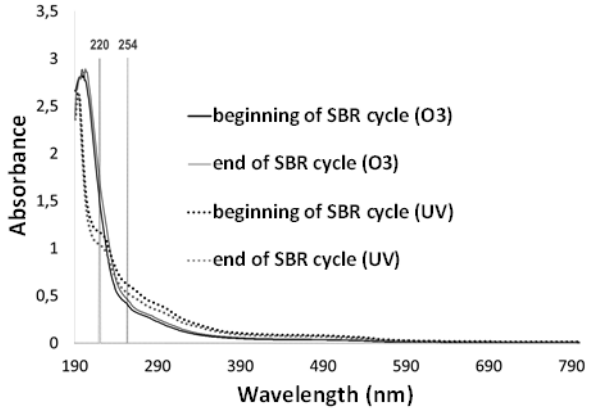

Figure 7. UV-vis spectra of effluent recirculated to SBR at the beginning and end of the biological cycle after AOP processes (ozonation - O3; UV irradiation - UV).

Also, no significant changes were detected through HPLC analysis on samples before and after the biological treatment of AOP-treated effluents. The ozonated effluent chromatograms showed the concentration of the residual amine 4A1NS and the area of the peak at 3.3 min essentially unchanged. The chromatographic profile of the UV-irradiated effluent also remained the same, with almost no peaks in any case. This limited performance of the biological treatment on AOP-processed effluents is possibly also due to the very low substrate concentrations at the start of the cycle, namely 127 and 116 mgCOD/L for the ozonized and irradiated effluents, respectively. In an industrial water resources conservation context, adsorption and/or membrane filtration operations could be used to produce water for reuse in manufacturing operations, either before or after the application of AOP. In these cases, AOP and subsequent biological polishing could be applied to more concentrated streams (the concentrates from the water recovery operations), favouring process kinetics and possibly improving COD removal performance.

CONCLUSIONS

The conversion of the recalcitrant 4A1NS amine present in an AGS-SBR biologically treated textile effluent was successfully achieved using the two AOP tested. A removal efficiency of 85% for this amine could be obtained with either 5 min of ozonation or 20 min of UV irradiation. In both cases, the UV-vis spectral profile and the HPLC chromatographic profile of the effluent were markedly altered, more extensive absorbance and peak area reductions being obtained with UV irradiation. COD removal in both AOP treatments was insignificant, but a simulated biological polishing using the AGS-SBR biomass and a 5-h SBR reaction cycle achieved COD removal yields of 15 and 20%, for irradiated and ozonated effluents, respectively. However, a small change in UV-vis spectral

profile indicating possible bioremoval of aromatic solutes was only observed for the effluent previously subjected to UV irradiation.

It can be concluded that ozonation and UV irradiation are promising options as polishing treatments for the effluents coming from AGS-SBR treatment of textile wastewaters, aiming to eliminate recalcitrant aromatic amines resulting from azo dye bioreduction. The recirculation of the AOP-treated effluent to the biotreatment stage was found to be a possible option to further reduce both COD and the dissemination of dye-derived pollutants in receiving water bodies or water reuse applications.

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