

TEXTILE DYEING WASTEWATER TREATMENT BY COMBINATION OF COUPLED AOPs AND AEROBIC SEQUENCING BATCH REACTOR

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Abstract - This study investigated the potential of combining coupled advanced Oxidation Processes (AOPs) and biological process for the treatment of real textile dyeing wastewaters. The samples were collected from a fabric dyeing industry in Perundurai SIPCOT. The colour absorbance of the wastewater ranged between 0.735-0.903 @ 436mm, 0.462-0.792 @ 525mm and 0.604-0.312 @ 620mm. the COD and BOD of the effluents were found to be in the range of 825-1350mg/L and 260-375mg/L. Samples were alkaline in nature with a low biodegradability ratio in the range of 0.22-0.31. coupled AOP treatments were conducted in a lab scale photo reactor of 6L capacity. Under the experimental conditions of ozone dosage 157mg/L, h₂o₂ dosage of 100mg/L and irradiation by UV-C lamp of 18W, the result obtained were 87-89% colour removal at 620mm, 38-42% COD reduction and improved biodegradability ratio 0.42-0.45 was achieved in 60minutes. The AOP treated wastewater was the subjected to biological degradation in a lab scale sequencing batch reactor of 5.7L capacity. However, it was found that treatment efficiency did not increase significantly even when the HRT, MLSS Concentration and SRT increases. Overall, AOP prior to biological treatment appears to be the best option regarding the treatment of real textile dyeing wastewater treatment.

Keywords: Coupled Advanced Oxidation Processes, Sequencing Batch Reactor, Biodegradation, Hydraulic Retention Time and Soilds Retention Time.

I. Introduction

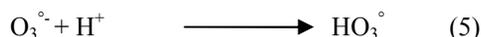
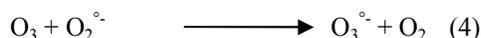
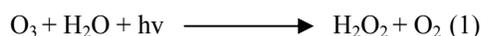
Textile industries are major consumers of water with high average water consumption and consequently one of the largest groups of industries causing intense water pollution [1]. In the last two decades, effluent discharges from textile dyeing industries poses a high threat for the environment in the Coimbatore, Erode and Tripur districts of Tamil Nadu [2]. The industries in these areas generate 80 million liters of effluents per day, these effluents are highly coloured and contain appreciable level of organic compounds. Moreover, absorbable organic halogens (AOX) are formed as a result of the use of bleaching chemicals [3]. When these effluent are discharged into the water course, the ecological consequences are degradation of surface water quality by reduction water transparency and sunlight penetration, thereby altering the photosynthetic activity [4]. In this frame, textile industry is confronted with the challenge of effective wastewater remediation. Thus decomposition of these organic dyes is of great significance to water purification and conservation [5].

Currently there are several different treatment methods applied to abate the textile wastewater pollution and can be divided into: i) physical and physico-chemical methods, ii) biological approaches and iii) chemical methods [6]. The most prominent drawback of most of the physico-chemical treatment processes (coagulation/flocculation, membrane processes or activated carbon adsorption and gas chlorination) is in secondary waste production (sludge), membrane fouling and costly adsorbent regeneration common practices, but they are also quite inefficient and

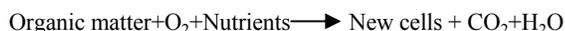
result in phase transfer but not in removal of pollutants [7]. Biological treatment methods on the other hand are considered to be inherently environmental friendly and cheaper compared to chemical ones [8]. The conventional biological processes which are in practice are aerated lagoons, trickling filters and activated sludge process. The major disadvantages of using the biological processes are no complete degradation of pollutants and the process requires long retention time [9].

To ease the stated problems, destructive treatment methods for the remediation of recalcitrant or hazardous pollutants are currently under investigation. By taking the advantages of the high oxidative power of the OH radical (2.8 V), which have the ability to oxidize and even mineralize the majority of organics in industrial effluents is used here to treat the recalcitrant pollutants [10]. Several advanced oxidation processes (AOPs) such as photocatalysis, Fenton and photo-Fenton has been used in the remediation of textile wastewater, and to make the textile effluent as biodegradable, AOPs are the best treatment technology [11]. In order to remove organic pollutants more effectively, the combination of powerful oxidants such as O₃ and H₂O₂ is a promising alternative. AOPs can degrade complex chemical structures to more easily degradable molecules, which can then be treated in a conventional and less expensive way eg: biological treatment. The use of coupled AOPs is an emerging new technology in recent years and some interesting coupled systems viz., Photofenton- ozonation (PFO), Photocatalytic ozonation (PCO), Sono-photocatalytic oxidation and O₃/UV/H₂O₂ have been proposed to treat various types of

industrial wastewater. In this study O₃/UV/H₂O₂ system was adopted, as the addition of H₂O₂ to the O₃/UV accelerates the decomposition of ozone which resulted in an increased rate of OH generation [12]. The literature studies showed that the ozonation based AOP is a promising method for primary treatment of textile wastewater as it removes appreciable level of colour and improves the biodegradability (<0.4) of the effluent [13]. Numerous works have been published in this area by Azam et al., 2012; Francese and Julia, 2014; Jose et al., 2012; Marija et al., 2014; Karthekeyan et al., 2011 and Sandip et al., 2013. The mechanisms of coupled AOPs (O₃/UV/H₂O₂) are shown below.



After improving the biodegradability of textile effluent, biological treatment capable of reducing the contaminant level of wastewater for many years can be adopted. Oxidation of organic matter in biological process may be aerobic or anaerobic process [10]. The equation (6) presented below represents the aerobic oxidation of organic matters in the presence of microorganisms [11].



In the present work, the combination of chemical oxidation using O₃/UV/H₂O₂ and biological degradation in SBR was applied to the treatment of a real textile effluent generated in the fabric dyeing unit. The feasibility of biological degradation by the biomass on the coupled AOPs treated textile effluent was investigated.

II. Materials and Methods

A. Textile wastewater collection and composition

Experiments were performed on real wastewater from textile factory located in perundurai, which is a dyeing unit where cotton and polyester fabrics are dyed using reactive dyes. The effluents were collected from equalization tank and stored in a cooling chamber at 4°C before use. The textile dyeing effluent is characterized mainly by high organic content, dyestuffs and salinity.

B. Chemicals and Reagents

Commercial H₂O₂ of (30%) were used to obtain OH[•] and all the chemicals used for study were obtained from the Merck chemicals. Concentrated sulphuric acid and sodium hydroxide solutions were used to achieve desired pH values in working solution. Deionized water was used to prepare all solutions.

C. Analyses

Analyses were performed in raw and treated wastewater (both AOP and biological) for colour (436nm, 525nm and 620nm), pH, COD, BOD, TDS and TSS according to the standard methods (APHA 2005) to follow the progress during the process. The decolourisation and degradation efficiencies were calculated by the following expressions.

$$\eta = ((A_0 - A_1) / A_1) \times 100 \quad (7)$$

$$\eta = ((C_0 - C_1) / C_1) \times 100 \quad (8)$$

Where, A₀ and A₁ are the absorbance of samples before treatment and after treatment. C₀ and C₁ are the concentrations of COD or BOD before treatment and after treatment (mg/L).

D. AOP reactor and Procedure

The AOP experiments were performed in a closed plexiglass box photoreactor of 6L capacity. Then pre-established amounts of hydrogen peroxide 30% (100mg/L) were added to the effluent to promote the generation of OH[•] and fed through inlet nozzle of the reactor. A 18W Philips mercury vapour lamp, located horizontally above the reactor which basically emits 254nm were used as artificial light source. O₃ was generated by O₃ generator (10g/h, Faraday ozone) and fed through the bottom of the reactor by means of diffuser. The O₃ input into the solution was 157mg/L, unreacted excess O₃ from the reactor was trapped and measured by iodometric titration as per standard methods. Periodically, samples were taken from the reactor and remaining H₂O₂ was eliminated by adding a small, but in excess, amount of sodium sulphite, in order to stop the reaction (as confirmed by preliminary experiments at different sulphite doses). Subsequently, samples were analysed for COD, BOD and absorbance (436nm, 525nm and 620nm) to observe the decolourization and degradation rate. A photographic view of a lab-scale reactor for coupled AOP is illustrated in Figure 1.

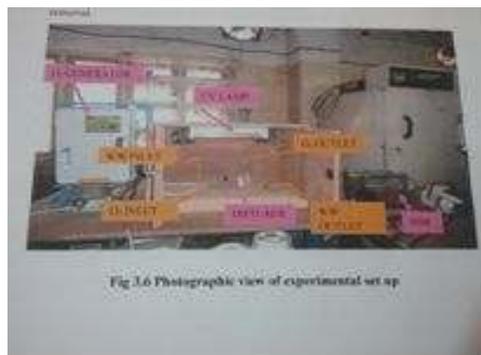


Fig.1.6 Photographic view of experimental set up

Figure 1: AOP reactor setup

E. Acclimation of activated sludge in Sequencing Batch Reactor and Procedure

The biological treatment system was composed of a 5.7L (plexiglass) aerobic lab-scale SBR, equipped with air

diffusers which provided the oxygen necessary to keep the aerobic condition. The reactor was seeded with biomass obtained from domestic wastewater treatment plant, to achieve an initial MLSS concentration of approximately 4000mg/L. In order to allow the biomass to get acclimatized to the AOP treated textile wastewater, a step-by-step feeding approach (10d) was followed during acclimation. The AOPs treated wastewater contains active hydroxyl radical, to stop their reactions during the biological process, sodium sulphite was added until the DO falls to zero. The excess sulphite was removed by aeration. The SBR was operated in fill, react, settle and draw mode in the ratio of (1:5:1:1) to constitute a cycle time of 8h. the fill and draw SBR procedures were operated in manual mode. Aeration was then shut down for 1h, during which the sedimentation stage proceeded. After the sludge was fully settled, the supernatant was removed from the reactor(decant time = 1h). SRT play a crucial role in the overall stability and biodegradation performance of a reactor. SRT control involved wasting an appropriate amount of MLSS on the daily basis. The concentration of dissolved oxygen was kept not lower than 3mg/L. MLSS concentration and pH were monitored throughout the experiments. Solution pH was adjusted to around neutral mark by adding a concentrated solution of sulphuric acid, prior to feeding to the bioreactors, in order to minimize any potential toxic and/or inhibitory effects on the biomass. Suitable proportions of essential biological nutrients (MgSO₄, CaCl₂, FeCl₂ and phosphate buffer) were also added to the solution. The samples were collected and analysed for colour, COD and BOD removal. The SBR set up is shown in figure 2. The experiments were conducted by changing one variable at a period of time, while keeping other parameters constant.

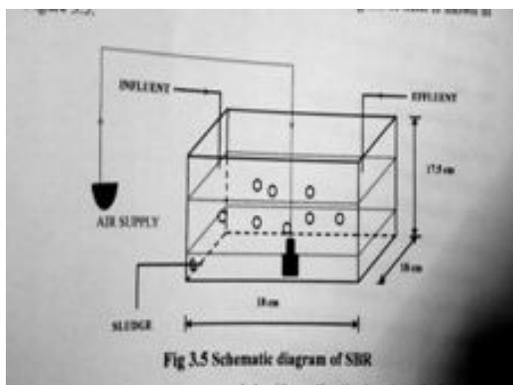


Figure 2 SBR setup

III. Results and Discussions

A. Characterisation of Real Textile Dyeing Effluent

Textile effluent was characterized as shown in table 1. The major pollutants identified in textile dyeing effluent can be summarized as colour, pH, organic load and salt.

TABLE I. Characterisation of Textile Dyeing Wastewater

PARAMETER	IWW 1	IWW 2	IWW 3
Color to naked eye	Black	Dark pink	Green
Color	436nm	0.735	0.903
	525nm	0.462	0.792
	620nm	0.312	0.604
pH	8.2	9.7	9.3
TDS (mg/L)	7140	6420	6260
TSS (mg/L)	285	230	200
COD (mg/L)	1350	826	941
BOD (mg/L)	378	260	270

The effluents were alkaline in nature, with pH varying from 8.2 to 9.7. The colour absorbance of the wastewater ranged between 0.735 to 0.903 at 436nm, 0.462 to 0.792 at 520nm and 0.312 to 0.580 at 620nm. The BOD₅ to COD ratios were in the range of 0.22 to 0.31, which means a poorly biodegradable sample. The wastewater had high TDS and TSS due to the addition of salts during dyeing process.

B. Coupled AOPs Treatment

With a view to ascertain decolourization and to improve the biodegradability of real textile dyeing wastewater coupled AOPs were used to give the pre-treatment. Ozone is usually coupled with another oxidant like H₂O₂ or UV irradiation to enhance the formation of OH[·] in aqueous phase. Many researchers found that alkaline nature of the sample helps in dissociation of H₂O₂ into HO⁻² at a high rate which in turn initiates O₃ decomposition more effectively than OH[·]. The results of the experimental run were depicted in figure 3 and 4. In this study, colour, COD and BOD were reduced by 87% - 89%, 38% - 43% and 11%-19% in 1 h and to 93-94%, 43-46% and 14-21% in 2h respectively. The BOD/COD ratio, which is an indicator of biodegradability was initially 0.22-0.31 but rose to 0.40-0.44 in 1h and to 0.42-0.45 in 2h, indicating an increase in biodegradability after AOP treatment. These results were also observed by Jose et al., 2012 and Aleksandra et al., 2010.

The result obtained in this study illustrates that degradation was slower than alterations that took place in chromophore groups, which led to colour removal. This indicates that the intermediate products formed were more resistant to hydroxyl radical attack than the parent compound during O₃/UV/H₂O₂ degradation. This low COD and BOD removal indicate that this effluent contained molecules that are very resistant to photochemical oxidation. In contrast to COD and BOD reduction, colour removal was very high (87-89%), this result shows that the AOP has a great potential for eliminating colour in textile

effluent. The literature shows that decolourisation and degradation efficiencies are mainly dependent on dye/organic matter contents, pH, temperature and ozone transfer to the wastewater solution, intensity of UV radiation and other factors.

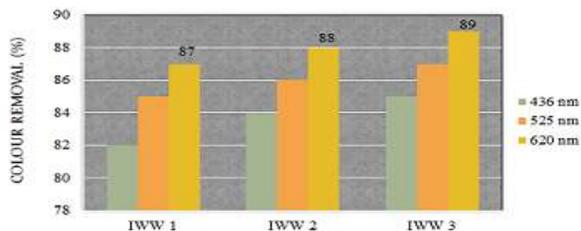


Figure 3 Colour Removals by coupled AOPs treatment

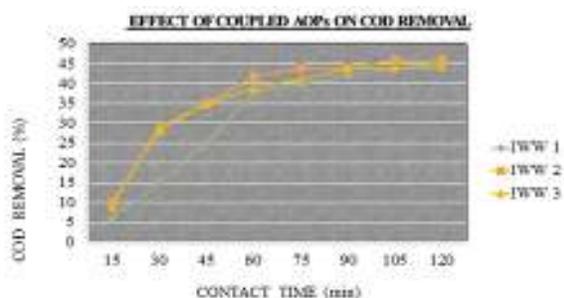


Figure 4 COD removal by coupled AOPs Treatment

A combination of the AOPs and biological processes could increase efficiency and reduce treatment costs for effluents with similar characteristics. Initial AOP application followed by biological processes to remove or reduce biorecalcitrant, non-biodegradable, and/or toxic pollutants has become a very common treatment for many types of complex matrices (Carmen et al., 2009; Jose et al., 2013; Adriana et al., 2012).

A. Performance of SBR

For most industrial wastewater, an acclimation period is necessary in order to gradually expose the microbial community to the potentially inhibitory or toxic organic compounds present. This allows for the development of appropriate enzyme-producing genes that are essential to induce biodegradation. In this study, the feeding pattern followed (described in section 2.5) appeared to be successful, since the targeted minimum COD removal level of 70% was achieved within 5 days of operation. The ability of properly acclimatized biomass to degrade non-easily biodegradable compounds has been frequently observed in the literature. Furthermore, fluctuation in the MLSS content during acclimation was minimal in all reactors, with an average value of 4000 ± 100 mg/L, which provides additional evidence of the stability of the process.

The treatment effectiveness in terms of removal efficiencies is reported in table 2. The data reported in table 2, shows that combined treatment (coupled AOPs followed biological process) was able to remove 91-92%, 85-88% and 89-94% of colour, COD and BOD respectively (HRT=5h). it is interesting to note that although the HRT increases from 5 to 21h, colour removal did not increase significantly which was not surprising because the effluent had already been treated by AOP, which removed a great deal of colours. Previous studies have reported that the decolourization of dyed effluent can be achieved by microorganisms and can be accomplished in two ways: either by bioaccumulation, in which dyes are adsorbed on the cell wall of microorganisms and or by biodegradation, in which dyes are oxidised by the specific enzyme system of the microorganisms. The COD removal is due to the aerobic biological treatment, which involves stabilization of biodegradable organic content of wastewater by the mixed population of microorganisms. The mixed aerobic microbial consortium used the organic carbon present in the effluent as its energy sources. The complex organic finally get converted to microbial biomass (sludge) and CO₂. From the table 2 it was observed that increasing the HRT did not seem to influence the decolourisation and degradation rate, and also longer HRT resulted in increase of operating cost. Therefore HRT was limited for a time period of 5h. the removal efficiencies were almost similar to those reported in the literatures.

TABLE II. Effect of HRT

HRT(h)	Colour removal (%)	COD removal (%)	BOD removal (%)
5	91-92	85-88	75-89
9	93-94	86-90	82-93
21	95-96	88-91	90-95

The influence of MLSS concentration on the effectiveness of SBR process for degradation is shown in figure 5. The maximum degradation was achieved at MLSS concentration of 4000mg/L (HRT=5H). The figure illustrates that higher concentration of MLSS promoted the SBR performance on organic removal. And also, no significant difference in COD removal efficiency was observed when the biomass was increased from 4000 to 5000mg/L. Hence optimum MLSS of 4000mg/L was selected. Beenish et al., 2013 suggested that increase in MLSS concentration increase the COD degradation rate. Several studies have demonstrated partial or complete degradation of dyes by pure and mixed cultures of bacteria. In many bio-treatment systems, mixed bacterial cultures have proved to be superior to single pure cultures achieving higher degree of dye degradation.

The effect of operation cycle per day on the degradation by SBR process is shown in figure 6. From the

figure it was observed that as the cycle increases, the degradation rate decreases. COD removal was 84-85% (2cycles). This indicated that SBR performance may be affected by the length of idle period. The idle time was 16,8 and 0 h for 1, 2 and 3 cycles per day, respectively. Therefore, sufficient idle period should be used to ensure the effluent quality. Based on the experimental data, 2 operating cycles per day was the optimal condition for the SBR.

The findings from SBR process suggest that there was no apparent advantage in increasing the SRT beyond 10d as the COD removal decreased with increase of SRT to 15d. The SRT at 10days gives maximum COD removal of 85-86%. The results are shown in figure 7. The process may fail if the design mean cell residence time is less than the maximum generation time of biomass (mainly bacteria) (Karia and Christian, 2009). Whereas, higher SRT deteriorates the quality of effluents and also causes operational problems such as poor settling, increased foaming and difficulty in supplying oxygen. Hence, the optimal SRT of 10d was selected.

IV. Conclusion

The result suggests that the use of coupled AOPs could be a feasible technique for decolourisation and biodegradation of textile dyeing wastewater and it could also become a cost effective technique at industrial scale application. It was observed that biological treatment in properly acclimatized SBR system could lead to a high organic matter removal as well as to a modest reduction in colour. In summary, the optimal reaction conditions to degrade this AOP treated textile wastewater were HRT=5h, MLSS concentration = 4000mg/L, Operation cycle per day = 2 cycles and

SRT = 10d. under these conditions, it was possible to reduce the colour(92%), COD (92%) and BOD (85%). This illustrates that the combination of the coupled AOPs and biological process is a viable method for treating coloured textile effluent.

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