TREATMENT OF TEXTILE WASTEWATER BY ADVANCED OXIDATION PROCESSES – A REVIEW

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ABSTRACT

The use of conventional textile wastewater treatment processes becomes drastically challenged to environmental engineers with increasing more and more restrictive effluent quality by water authorities. Conventional treatment such as biological treatment discharges will no longer be tolerated as 53% of 87 colours are identified as non-biodegradable. Advanced oxidation processes hold great promise to provide alternative for better treatment and protection of environment, thus are reviewed in this paper. An overview of basis and treatment efficiency for different AOPs are considered and presented according to their specific features.

KEYWORDS: H_2O_2/UV ; O_3/UV ; O_3/H_2O_2 ; $O_3/H_2O_2/UV$

INTRODUCTION

Textile wastewater includes a large variety of dyes and chemicals additions that make the environmental challenge for textile industry not only as liquid waste but also in its chemical composition (Venceslau et al., 1994). Main pollution in textile wastewater came from dyeing and finishing processes. These processes require the input of a wide range of chemicals and which dyestuffs, generally are organic compounds of complex structure. Because all of them are not contained in the final product, became waste and caused disposal problems. Major pollutants in textile wastewaters are highsuspended solids, chemical oxygen demand, heat, colour, acidity, and other soluble

substances (Dae-Hee *et al.*, 1999). The removal of colour from textile industry and dyestuff manufacturing industry wastewaters represents a major environmental concern. In addition, only 47% of 87 of dyestuff are biodegradable (Pagga and Brown, 1986). It has been documented that residual colour is usually due to insoluble dyes which have low biodegradability as reactive blue 21, direct blue 80 and vat violet with COD/BOD ratio of 59.0, 17.7, and 10.8, respectively (Marmagne and Coste, 1996). Conventional oxidation treatment have found difficulty to oxidize dyestuffs and complex

difficulty to oxidize dyestuffs and complex structure of organic compounds at low concentration or if they are especially refractory to the oxidants. To ease the stated problems advanced oxidation processes (AOP_s) have been developed to generate hydroxyl free radicals by different techniques. AOP_s processes are combination of ozone (O₃), hydrogen peroxide (H₂O₂) and UV irradiation, which showed the greatest promise to treat textile wastewater. These oxidants effectively decolorized dyes, however did not remove COD completely (Ahmet *et al.*, 2003; Lidia *et al.*, 2001; Stanislaw *et al.*, 2001; Tzitzi *et al.*, 1994).

Textile Wastewater Characteristics

Composite textile wastewater is characterized mainly by measurements of biochemical oxygen demand (BOD), chemical oxygen demand (COD), suspended solids (SS) and dissolved solids (DS). Typical characteristics of textile industry wastewater are presented in Table 1. Results in Table 1 show a large extent of variation from plant-to-plant and sample-tosample. As presented in Table 1, COD values of composite wastewater are extremely high compare to other parameter. In most cases BOD/COD ratio of the composite textile wastewater is around 0.25 that implies that the wastewater contains large amount of nonbiodegradable organic matter.

Advanced Oxidation Process (AOPs)

The goal of any AOPs design is to generate and use hydroxyl free radical (HO[']) as strong oxidant to destroy compound that can not be oxidized by conventional oxidant. Table 2 shows the relative oxidation potentials of several chemical oxidizers. Advanced oxidation processes are characterized by production of OH radicals and selectivity of attack which is a useful attribute for an oxidant. The versatility of AOP is also enhanced by the fact that they offer different possible ways for OH' radicals. A list of the different possibilities offered by AOP is given in Table 3. Generation of HO is commonly accelerated by combining O₃, H₂O₂, TiO₂, UV electron-beam irradiation radiation. and ultrasound. Of these, O₃/H₂O₂, O₃/UV and H₂O₂/UV hold the greatest promise to oxidize textile wastewater.

Table 1. Composite textile industry wastewater characteristics*

Parameters	Values
рН	7.0-9.0
Biochemical Oxygen Demand (mg/L)	80 - 6,000
Chemical Oxygen Demand (mg/L)	150 - 12,000
Total Suspended Solids (mg/L)	15 - 8,000
Total Dissolved Solids (mg/L)	2,900 - 3,100
Chloride (mg/L)	1000 - 1600
Total Kjeldahl Nitrogen (mg/L)	70 - 80
Colour (Pt-Co)	50-2500

^{*}Sheng and Chi, 1993; Tzitzi *et al.*, 1994; Venceslau *et al.*, 1994; Altinbas *et al.*, 1995; Olcay *et al.*, 1996; StanisŁaw and Monika, 1999; Gianluca and Nicola, 2001; Arslan and Isil, 2002; Arslan *et al.*, 2002; Cleaner Production Program-CPP, 2002; Georgiou *et al.*, 2002; Mehmet and Hasan, 2002; Ahmet *et al.*, 2003; Metcalf and Eddy, 2003; Azbar *et al.*, 2004)

Table 2.	Oxidizing	potential f	for conven	tional o	xidizing	agents*
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	Electrochemical	EOP relative to
Oxidizing agent	oxidation potential	chorine
	(EOP), V	
Fluorine	3.06	2.25
Hydroxyl radical	2.80	2.05
Oxygen (atomic)	2.42	1.78
Ozone	2.08	1.52
Hydrogen peroxide	1.78	1.30
Hypochlorite	1.49	1.10
Chlorine	1.36	1.00
Chlorine dioxide	1.27	0.93
Oxygen (molecular)	1.23	0.90

*(Carey, 1992; Techcommentary, 1996; Zhou and Smith 2002; Metcalf and Eddy, 2003)

Table 3. Advanced oxidation processes

$H_2O_2/UV/Fe^{2+}$ (photo assisted Fenton)			
H_2O_2/Fe^{2+} (Fenton)			
Ozone/UV(also applicable in the gas phase)			
Ozone/H ₂ O ₂			
Ozone $/UV/H_2O_2$			
Ozone/TiO ₂ /Electron-beam irradiation			
Ozone/TiO ₂ /H ₂ O ₂			
Ozone + electron-beam irradiation			
Ozone/ultrasonics			
H_2O_2/UV			

ULTRAVIOLET LAMP

Ultraviolet light is part of the light spectrum. The process usually involves the use of low-pressure UV lamps with a principal wavelength of 254 nm. Since the maximum absorption of ozone molecules is at 253.7 nm, the light source commonly used is a medium-pressure mercury lamp wrapped in a quartz sleeve that can generate the UV light at wavelength of 200-280 (Zhou and Smith, 2002). Application of UV lamp for textile wastewater treatment has been reported by StanisŁaw and Monika (1999). They have applied two different UV radiations; 150W, λ = 254-578 nm and 15W, λ = 254 nm, to the synthetic textile wastewater for 1 to 3 h. They have recorded significant reduction (47 to 30%) in microbial inhibitory action for optimum radiation time of 1 hour.

OZONE

Ozone is a powerful oxidant agent for water and wastewater. Once dissolved in water, ozone reacts with a great number of organic compounds in two different ways: by direct oxidation as molecular ozone or by indirect reaction through formation of secondary oxidants like hydroxyl radical (Baig and Liechti, 2001). The conventional fine bubble contactor is the most widely ozone generator used because of the high ozone transfer efficiency (90%) and high performance (Zhou and Smith, 2002). Ozone application can be generalized into two; a powerful disinfection and a strong oxidant to remove colour and odour, eliminating trace toxic synthetic organic compounds and assisting in coagulation (Langlais et al., 1991).

According to Mehmet and Hasan (2002), mg/dm^3) ozonation (300 increased the biodegradability index of textile wastewater by 1.6 times. Jianging and Tingwei (2001) documented 11-66 times increase in biodegradability index for wastewater containing azo dye, while this increment reached to 80 times for wastewater containing simulated reactive dye and reactive yellow 84 (Koch *et al.*, 2002). These findings revealed that increase in biodegradability index was influenced by type and concentration of dye.

Results presented by a few researchers revealed that ozone decolorize all dyes, except nonsoluble disperse and vat dyes which react slowly and take longer time (Namboodri et al., 1994; Marmagne and Coste, 1996; Rajeswari, 2000). Furthermore, it has been documented that colour removal using ozonation from textile wastewater is depended on dye concentration (Sheng and Chi, 1993, Mehmet and Hasan, 2002, Konsowa 2003). Higher initial dye concentration of textile wastewater causes more ozone consumption. Increasing ozone concentration enhances mass transfer that causes an increase in ozone concentration in liquid phase, which increase colour removal. The other possible explanation is that more intermediates, which consume more ozone are generated when the initial dye concentration is high. According to Ahmet et al. (2003) and Arslan and Isil (2002), 40-60 min ozonation of biotreated textile wastewater yielded 99% of decolorization efficiency. Gianluca and Nicola (2001) documented colour removal of biotreated textile wastewater was depended on initial COD of textile wastewater. About, 99% and 95% of colour removal was achieved when COD initial was 160 and 203 mg/L, respectively. In addition, colour removal efficiency increased with increasing the temperature from 25°C to 50°C (Rajeswari, 2000).

Previous researchers documented that the rate of dye oxidation increased slightly with increasing solution pH (Mehmet and Hasan, 2002; Konsowa, 2003; Azbar et al., 2004). Arslan et al., (2002) documented that high colour removal of simulated reactive dye bath effluent was achieved at pH 7 when using ozone concentration of 2970 mg/L and remained unchanged at pH 11. Moreover, According to Koch et al. (2002) ozonation of the hydrolyzed dye (Reactive Yellow 84) decreased as solution pH decreased from 6.1 to 3.2 when ozone concentration was 18.5 mg/L. Konsowa (2003) documented 32% reduction in the dve decolorization time when the pH values changed from 2 to 12. Higher colour removal at alkaline pH could be due to enhancement of ozone decomposition by hydroxyl radical at alkaline pH values. The rate of ozone decomposition is

favored by the formation of hydroxyl radicals at higher pH values. The reaction pH had to be at least 7 to enhance ozone decomposition and high colour removal.

Previous researchers documented that COD and TOC removals using ozonations are dependent on pH. Lidia et al. (2001) reported low COD removal of disperse dyes (10%) at pH 8 using 0.5g dm⁻³ of ozone concentration, while, Arslan et al. (2002) documented high TOC removal for simulated reactive dye bath effluent at pH 7 when using ozone concentration of 2970 mg/L. At pH 7, the radical type reaction becomes effective and simultaneously the inhibiting effect of carbonate ions is not yet very pronounced. Thus higher TOC and COD removals could be expected. Furthermore, Rajeswari (2000)documented that TOC and COD removals efficiency was dependent on temperature. He recorded that TOC and COD removals increased with increasing the temperature from 25°C to 50°C. Koch et al. (2002) documented TOC removal of hydrolyzed reactive dye was less than COD throughout the ozonation process and it was 30% and 25% after 60 and 90 min with an ozone concentration of 18.5 and 9.1 mg/l, respectively.

Sheng and Chi (1993) documented COD removal depended on strength of dye waste, where COD reduction was slight from the medium and high dye waste. The low COD reduction is attributable to the fact that the structured polymer dye molecules are oxidized by ozonation to small molecules, such as acetic acid, aldehyde, ketones, etc., instead of CO₂ and water. Thus considerable amount of COD is attributed to these small organic molecules. According to Gianluca and Nicola (2001), COD removal of biotreated textile wastewater was depended on initial COD of textile wastewater. About 67% and 39% of COD removal was achieved when COD initial was 160 and 203 mg/L, respectively. According to Koch et al. (2002), COD removal throughout ozonation process was 50% and 40% after 60 and 90 min with an ozone concentration of 18.5 and 9.1 mg/l, respectively.

Taken together, high colour removal can be achieved on wastewater, which contain high initial dye concentration and low initial COD. Alkaline pH and high temperature were also found as favorable conditions for high TOC and COD removals. In spite of having high colour removal efficiency limited COD and TOC removal efficiencies were obtained. This could be explained by incomplete oxidation of organic materials and production of small organic molecular fragment along with destruction of dyestuff that not being completely oxidized.

O₃/UV

(2001), According to Rein conventional ozonation of organic compounds does not completely oxidize organics to CO₂ and H₂O in many cases. Remaining intermediate products in some solution after oxidation may be as toxic as or even more toxic than initial compound and UV radiation could complete the oxidation reaction by supplement the reaction with it. UV lamp must have a maximum radiation output 254 nm for an efficient ozone photolysis. The O₃/UV process is more effective when the compounds of interest can be degraded through the absorption of the UV irradiation as well as through the reaction with hydroxyl radicals (Rein, 2001; Metcalf and Eddy, 2003). The O₃/UV process makes use of UV photons to activate ozone molecules, thereby facilitating the formation of hydroxyl radicals.

Hung-Yee and Ching-Rong (1995) documented O₃/UV as the most effective method for decolorizing of dyes comparing with UV oxidation by UV or ozonation alone. While, Perkowski and Kos (2003) reported no significant difference between ozonation and O₃/UV in terms of colour removal. Even though ozone can be photodecomposed into hydroxyl radicals to improve the degradation of organics, UV light is highly absorbed by dyes and very limited amount of free radical (HO') can be produced to decompose dyes. Thus same colour removal efficiencies using O₃ and O₃/UV could be expected. In normal cases, ozone itself will absorb UV light, competing with organic compounds for UV energy. However, O₃/UV treatment is recorded to be more effective compared to ozone alone, in terms of COD removal. Bes-Piá et al. (2003) documented that O₃/UV treatment of biologically treated textile wastewater reduced COD from 200-400 mg/L to 50 mg/L in 30 minutes, while, using ozone alone COD reduced to 286 mg/L in same duration. Azbar et al. (2004) documented that using O₃/UV process high COD removal would be achieved under basic conditions (pH=9).

H_2O_2/UV

Oxidization of the textile wastewater with hydrogen peroxide alone has been found ineffective at both acid and alkali values (Olcay *et al.*, 1996), while under UV irradiation, H_2O_2 are photolyzed to form two hydroxyl radicals (2OH') that react with organic contaminants (Crittenden *et al.*, 1999). Application of UV to synthetic textile wastewater for one hour with addition 2 ml/L of H_2O_2 decreased the inhibitory of microbial growth during subsequent biodegradation of textile wastewater form 47 to 26% (Stanislaw and Monika, 1999; Stanislaw; *et al.*, 2001).

Galindo and Kalt (1998) documented H_2O_2/UV process was more effective in an acid medium (pH \approx 3-4) in term of discolouration. Nonefficient colour removal at alkaline pH is contributed to the fact that hydrogen peroxide undergoes decomposition leading to dioxygen and water, rather than producing hydroxyl radicals under UV radiation. Therefore the instantaneous concentration in HO' is lower than expected. Furthermore, the H_2O_2/UV process is more sensitive to the scavenging effect of carbonate at higher pH values. Same authors reported that temperature did not have a significant effect on discolouration (Galindo and Kalt, 1998).

Others documented that removal of textile dye by H₂O₂/UV increased as doses of effective hydrogen peroxide increased up to a "critical" value (Galindo and Kalt, 1998; Arslan et al., 1999; Nilsun 1999). High concentration of the H₂O₂ acts as a radical scavenger, while; low concentration of H₂O₂ generates not enough of hydroxyl radicals (OH') that consumed by dye and this leads slow rate of oxidation. Thus a trade-off between them will result in an optimum H₂O₂ dose, which still needs to be verified experimentally. Arslan and Isil, (2001, 2002) also documented that treatment of textile wastewater was not effective via H₂O₂/UV-C oxidation unless a preliminary ozonation period was introduced to produce sufficient OH' to observe a significant colour and COD reduction. Previous researchers documented even at 50 mM of H₂O₂ concentration, raw textile wastewater did not suffer any significant oxidation and COD removal was negligible but COD removal efficiencies of the biotreated effluent was significant (Arslan and Isil, 2001; 2002).

Results presented by researchers revealed that sufficient reaction time for dye removal using H_2O_2/UV process was found to be subjective. Complete destruction of reactive dyes and azo dye have been recorded in 30-90 min by previous researches (Georgiou *et al.*, 2002; Mariana *et al.*, 2002; Rosario *et al.*, 2002; Tanja *et al.*, 2003), while, Percowski and Kos (2003) documented more than 99% colour reduction of dye house wastewater after 2 hours. Shyh-Fang *et al.* (1999) documented that the longer reaction time the more advantageous for colour removal where colour removal at 10 min was only 9% of removal at 120 min.

Georgiou et al. (2002) and Rosario et al. (2002), documented TOC removal of reactive dyes under H₂O₂/UV process up to 80-82% in 1-2 h. While, Arslan et al. (2002) found that TOC removal for simulated reactive dye bath effluent of H₂O₂/UV-C was only 30.4% and 13.9% using 680 mg/L of H₂O₂ at pH 3 and 7, respectively. Although, in some cases it has been reported that organic substances undergo photochemical reactions as a consequence of light absorption, rarely these transformations contribute to the TOC reduction. In this sense, the light absorbed by organic molecules can be generally considered as wasted light. Reduced efficiencies can also result during photolytic treatment of wastewaters containing suspended material, since a fraction of irradiated energy is scattered by these particles. Due to the fact that the inhibitory effect on treatment efficiency for the UV/H₂O₂ system is related directly to the decreasing light transmittance of the less diluted wastewater matrix, it can be inferred that for more concentrated effluents exhibiting higher optical density, light assisted AOPs are less suitable.

Galindo and Kalt (1998) documented the inverse relationship between the initial dye concentration and the efficiency of UV/H_2O_2 treatment. An increase in the dye concentration induces a rise of the internal optical density. Dyes react; consequently, the solution becomes more and more impermeable to UV radiation. Therefore, the rate of photolysis of hydrogen peroxide directly depends on the incident intensity. As a consequence, when initial dye concentration rises, the production of hydroxyl radicals decreases.

The relationship between UV light intensity and dye decomposition in UV/H₂O₂ process has been investigated by Shen and Wang (2002). The decomposition rate of dye was found to increase with increasing UV light intensity. They documented that more than 90% of the dye was decomposed at 82 Wm⁻². But for the UV light intensity higher than 102 Wm⁻², further increase of UV energy only slightly improved the decompostion efficiency of dye indicating the photons provided was excessive.

O₃/H₂O₂ (PEROXONE)

The addition of both hydrogen peroxide and accelerates ozone to wastewater the decomposition of ozone and enhances production of the hydroxyl radical. At acidic pH, H₂O₂ reacts only very slowly with O₃ whereas at pH values above 5 a strong acceleration of O₃ decomposition by H₂O₂ has been observed (Staehlin and Hoigne, 1982). At higher pH, even very small concentration of H₂O₂ will be dissociated into HO₂- ions that can initiate the ozone decomposition more effectively than OH⁻ ion (Staehlin and Hoigne, 1982; Glaze and Kang, 1989). Arslan et al. (1999) documented that H₂O₂/O₃ treatment of synthetic dyehouse highly depended on the pH of the effluent. They have documented 74% ozone absorption at pH 11.5 and 10 mM H_2O_2 whereas at the same concentration of H₂O₂ and pH 2.5, ozone absorption was only 11%. This phenomenon could be attributed to the that fact that the higher the pH, the more H_2O_2 will be dissociated into HO₂⁻ ions. As a result, the ozone decomposition rate will increase with increasing pH.

Complete decolourization of C.I. Reactive Blue 220 and C.I. Reactive Yellow 15 using H₂O₂/O₃ process is achieved in 90 min (Tanja et al., 2003). According to Tanja et al. (2003) C.I. Reactive Blue 220 was the most difficult to decolorize and C.I. Reactive Yellow 15 the easiest. On the other hand, Tanja et al. (2003) have documented that same dyes achieved decolourisation in 20 min when treated using H_2O_2/Fe^{2+} . This could be due to difference in medium pH. Decolourisation with H_2O_2/Fe^{2+} was performed in an acidic medium (pH=3), whilst for H_2O_2/O_3 in pH=12. Hydrogen peroxide in alkaline medium react with sodium hydroxide, as a result lower concentrations of hydrogen peroxide are available for the formation of hydroxyl radicals.

The inhibitory performance of H_2O_2/O_3 process on microbial growth depended on the H_2O_2 to O_3 mass ratio. This ratio ranged from 0.3 to 0.6 for different type of dye (Glaze, 1987; Singer and Reckhow, 1999; Rein, 2001). StanisŁaw and Monika (1999) documented 20% inhibitory effect of the optimum H_2O_2 (2 cm³) and O_3 (100 - 200 mg dm⁻³) on microbial growth.

$0_3/H_2O_2/UV$

The addition of H_2O_2 to the O_3/UV process accelerates the decomposition of ozone, which results in an increased rate of OH[•] generation (Teccommentary, 1996). Among all AOPs, for dye house wastewater and acetate, polyester fiber dying process effluent combination of $H_2O_2/O_3/UV$ appeared to be the most efficient in terms of decolouration (Azbar *et al.*, 2004; Perkowski and Kos, 2003).

Arslan and Isil (2001; 2002) reported that COD removal efficiency of raw textile wastewater increased from 18% to 27% by using sequential ozonation and H_2O_2/UV and in the case of biotreated textile effluent, a preliminary ozonation step increased COD removal of the H_2O_2/UV -C treatment system from 15% to 62%. Azbar *et al.* (2004) documented 99% COD removal from acetate and polyester fiber dying process effluent in batch mode operation using combination of $O_3/H_2O_2/UV$ in 90 min.

According to Arslan and Isil (2001; 2002), in sequential ozonation and H_2O_2/UV , TOC removal rate was accelerated from 14% (H_2O_2/UV -C) and 17% (O₃) to 50% by successive treatment of the raw textile effluent. In the case of biotreated textile effluent a preliminary ozonation step increased TOC removal from 0% to 34% but did not appear to be effective than applying a single ozonation process in terms of TOC abatement rates.

The inhibitory effect of O₃/UV/H₂O₂, when added $1 \text{ cm}^3 \text{ dm}^{-3} \text{ of } \text{H}_2\text{O}_2$ and $60 - 150 \text{ mg dm}^{-3}$ ozone on microbial growth during subsequent biodegradation of textile wastewater accounts for only 10%, while untreated wastewater exhibited 47% of inhibitory action. The optimal dose of using H_2O_2 alone at which the microbial growth was inhibited by only 30% was about 1.5cm³ dm⁻ ³. Increasing dose of H_2O_2 to 32 cm³dm⁻³ increased the inhibitory to 80% where the optimum ozone dose was about 100 mg dm⁻³ for effective biodegradation (StanisŁaw and Monika, 1999).

CONCLUSIONS

Advanced Oxidation Processes represent a powerful treatment for refractory and/or toxic pollutants in textile wastewaters. By properly combining ozone, hydrogen peroxide and UV different AOP techniques have been developed thus allowing to make choice the most appropriate for the specific problems. Taking into consideration that the efficiency of AOPs is compound specific, the final choice of the AOP system can be made only after preliminary laboratory tests.

There are many research needs to be done in field of AOP_S for textile wastewater to provide:

- Study the efficiency of different candidates' process under different controlled conditions.
- Identification of intermediates and byproducts and their toxicity.
- Study the sequences operation effect of the AOPs agents.
- Identification of scale-up parameters and criteria for cost effectiveness

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