Abstract review on:

Color Removal from textile effluents by biological processes

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May 2017

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Abstract

Dye decolorization and degradation is today one of the most important issues in textile facilities. The aesthetic, environmental and toxicity problems of discharging textile effluents in a water body should be taken into account to design a proper effluent treatment plant. Biological treatment, either aerobic or anaerobic, is generally considered to be the most effective means for the removal of the bulk of pollutants from complex or high-strength organic wastewater. This short review aims to report the effect of biological treatment in dyes removal, considering the aerobic and anaerobic treatments separately and combined. The most effective and feasible plant layout for color removal illustrated in the discussed.

1. Introduction

The textile industry is one of the most polluting industrial sectors. The production of textiles involves many different steps and most of these processes generate highly contaminated liquid streams (Cervantes, 2009). Textile wastewater usually contains refractory dyes and other organic chemicals causing a variety of problems in the natural aquatic environment due to its intense color and toxicity (Oh et al., 2004). Dye molecules consist of a chromophore component which generates the color, and fixed the dye into or within the fibers (Salleh et al. 2011). There are about 12 classes of chromogenic groups, the most common being the azo type, which makes up to 60-70% of all textile dyestuffs produced, followed by the anthraquinone type (Buckley et al. 1992; Carniell et al. 1995).

The biological treatment, either aerobic or anaerobic, is generally considered to be the most effective means for the removal of the main pollutants from complex or high-strength organic wastewater (Rai et al., 2005). Biological removal of the dyes from textile and dyestuff manufacturing industry can be broadly classified into three categories: aerobic treatment, anaerobic treatment and combination of them. On the other hand, only the biological process treatment of dye waste and textile effluents, depending on the local regulations, may be not sufficient (Carliell et al., 1995). Biological treatment using activated sludge in aerobic conditions is one of the regularly used treatment methods for textile dyeing effluents (Joshi and Purwar, 2004). The results of various researchers showed that dyestuff were not significant biodegradation rates under aerobic conditions (Chrisie, 2005). The conventional activated sludge process is not highly effective in the treatment of textile wastewater, regarding color, since most aerobic bacteria are incapable of completely degrading the dye molecules (Shaul et al., 1991). However, research is still on going to isolate aerobic microorganisms, such as fungi and bacteria, capable of degrading dyes and dyes related compounds (Neresh et al., 2013). Although some efforts to decolorize dyes under aerobic conditions have been successfully, the general perception of non-biodegradability of most azo dyes persists (Shore, 1995). In contrast, reductive cleavage of azo bonds readily occurs under anaerobic conditions (Field et al., 1995). It is reasonable to assume that a significant color removal may occur in anaerobic wastewater treatment systems (Cervantes, 2009). The anaerobic biodegradation process consists of a decolorization stage where the microorganism breaks the azo dye linkage of nitrogen double bond followed by a second stage which involves the degradation of the aromatic amines (Van der Zee, 2003). The limited past investigations have shown that azo dyes can be completely decolorized (O'Neill et al., 2000). The successful application of anaerobic technology for the treatment of industrial wastewaters depends on the development of high-rate bioreactors, which achieve a high reaction rate per unit reactor volume by retaining the biomass in the reactor for long periods of time (Naimabadi et al., 2009). On the other hand, anaerobic decolorization followed by aerobic post-treatment is generally recommended for treating colored wastewater from textile and dyestuff manufacturing industries (Brown, 1987). This condition can be implemented either by spatial separation

of the anaerobic and aerobic sludge using a sequential anaerobic–aerobic reactor system or within one reactor, commonly termed an integrated anaerobic–aerobic reactor system (Field et al., 1995; Zitomer et al., 1993; Van der Zee and Villaverde, 2005).
The aim of this review is to report literature studies of dyes removal under aerobic and anaerobic conditions and compare them in terms of decolorization efficiency achieved, and possible full scale industrial applications.

2. Aerobic treatment

Bacteria and fungi are the type of microorganisms more commonly used for the decolorization of dyes under aerobic conditions. Bacteria are able to grow faster than fungi (Balamurugan et al., 2011). Aerobic bacteria capable of degrading various dyes have been used more than three decades. Numerous bacteria capable to degrade dyes have been reported. Efforts to isolate bacterial cultures capable of degrading azo dyes started in 1970's with reports of *Bacillus subtilis*, then *Aeromonas hydrophilia,* followed by *Bacillus cereus* (Anjaneyulu et al., 2005). Aerobic removal of azo compounds by *Aeromonas hydrophilia* was reported (Idaka et al., 1987). Bacterium *Pseudomonas luteola* was isolated from the sludge of an activated sludge treatment system treating dyeing wastewater, which removed the color of four reactive azo dyes (Hu, 1994). Some researchers have been also applied sulfate-reducing bacteria for degradation of dyes (Parshetti et al., 2009; Togo et al., 2008). During the past years, many bacterial strains, that can aerobically decolorize azo dyes, have been isolated as well (Naresh et al., 2013). Many bacterial strains have shown to decolorize dyes, such as *Bacillus megaterium, Alcaligenes faecalis 6132, Rhodococcus erythropolis 24, Bacillus licheniformis LS04, Rahnella aquatilis, Acinetobacter guillouiae, Microvirgule aerodenitrificans, Pseudomonas desmolyticum NCIM 2112* (Han et al., 2012; Kalme et al., 2007; Praveen Kumar et al., 2012). In nature, the most efficient class of microorganisms for the degradation of synthetic dyes is the white-rot fungi. White-rot fungi are a class of microorganisms that produce enzymes capable of decomposing dyes under aerobic conditions (Ali, 2010). White-rot fungi such as *Dichomitus squalens, Daedalea flavida, Irpex flavus and Polyporus sanguineus* have been used widely in decolorization and degradation of textile wastewaster of many chromophoric groups of dyes (Chander et al., 2007). Other fungi such as, *Hirschioporus larincinus, Inonotus hispidus, Phlebia tremellosa* and *Coriolus versicolor* have been shown to decolorize dye-containing effluent (Banat et al., 1996; Kirby, 1999). White rot cultures have been used for the removal of azo dyes (Holkar et al., 2016). Bacteria and fungi strains commonly used in the biodegradation of textile dyes are listed in Table 1.

Table 1: Bacteria and Fungi strains commonly used in dye biodegradation

Culture	Dye	Dye removal (%)	References
P. chrysosporium fungi	Coracryl violet	100	Chander et al., 2007
P. chrysosporium fungi	Coracryl pink	100	
D. squalens fungi	Coracryl pink	100	
P. sanguineus fungi	Coracryl black	67	
D. flavida fungi	Coracryl pink	53	
T. versicolor ATCC 20869	Remazol blue	98	Toh et al., 2003
P. chrysosporium ATCC 24725	Remazol red	97	
P. chrysosporium ATCC 24725	Remazol blue	95	
Trametes species CNPR 4783	Remazol blue	89	
T. versicolor ATCC 20869	Remazol red	85	
Trametes species CNPR 4801	Remazol blue	58	

Aspergillus niger fungi	Scarlet Red	80	Kousar et al., 2000
Bacteria consortium SKB-II	Congo red	90	Tony et al., 2009
Bacteria consortium SKB-I	Blue BCC	74	
T. versicolor DSM 11269	Disperse red I	50	Junghanns et al., 2006
Myrioconium sp. UHH 1-6-18-4	Disperse blue 1	43	
S. rugosoannullata DSM 11372	Reactive red 4	31	

The use of bacteria and fungi for the complete decolorization and degradation of dyes from textile effluent has the advantages of being a low-cost process and the ability to complete the mineralization of dyes with nontoxic by-products (Dawkar et al., 2010). However, degradation of dyes in the textile wastewater by white-rot fungi has some intrinsic disadvantages, such as the long growth phase and the requirement of nitrogen restrictive environments, unreliable enzyme production and large reactor size due to the long holding time (14 days) for complete degradation (Anastasi et al., 2011). This process is not applicable to full-scale textile wastewater treatments because it is a very slow process and provides a suitable environment for the growth of autochthonous microorganisms (Toor et al., 2010).

3. Anaerobic treatment

Anaerobic decolorization involves an oxidation-reduction reaction with hydrogen rather than free molecular oxygen in aerobic system allowing azo and other water-soluble dyes to be decolorized (Robinson et al., 2001). Anaerobic reduction of azo dyes can be an effective and economic treatment process for color removal from textile wastewater (Erkurt, 2010).

The efficacy of various anaerobic treatment applications for the degradation of a wide variety of dyes has been demonstrated repeatedly (Forgacs et al., 2004). Many anaerobic bacteria can decolorize azo dyes under anaerobic conditions via reduction of the azo bond (Chrisie, 2005). Anaerobic discoloration of textile effluents is not yet well established, although successful pilot-scale and full-scale plants have been reported (Quezada et al. 2000; Tan et al. 2000; Willetts et al., 2000). Decolorization of the mono azo dye C.I. Acid orange 7 was reported by Van der Zee et al. (2000). In different studies, several high rate anaerobic reactors such as UASB and ABR were used in textile wastewater treatment (Sen et al., 2003). Applicability of a thermophilic UASB anaerobic system as a unit operation for the discoloration of synthetic textile dye wastewater clearly indicates that it has a significant advantage over an equivalent mesophilic system, and can effectively decolorize such a wastewater with a higher efficiency (Willetts et al. 2000). Discoloration of commercially relevant azo dye, Orange II, Black 2 HN, under anaerobic conditions, using a sequencing batch biofilter, showed > 99% color removal up to a dye concentration of 400 mg/l for both dyes (Manu & Chaudhari 2002). Among the different reactor types studied, the anaerobic filter and the UASB reactor gave good color removal efficiencies. A major advantage of this anaerobic system, apart from the discoloration of soluble dyes, is the production of biogas (Sen et al., 2003). Table 2 shows some literature results for dyes removal percentage under anaerobic conditions.

Table 2: Color removal efficiency of different anaerobic systems

Dye	Reactor	HRT	Color removal (%)	Reference
Acid red 42 (azo) Direct red 80 (azo) Disperse Blue 56 (anthraquinone)	UASB	16h	62% 81% 0%	(Gonçalves, 1993)
Acid Orange 7 (azo) Acid Orange 8 (azo) Acid Orange 10 (azo) Acid Red 14 (azo)	Fluidized bed	24h 12h 12h 24h	90% 98% 81% 86%	(Seshadri et al., 1994)
Acid Yellow 17 (azo) Basic Blue 3 (phenoxazine)	UASB	8-20 h	20% 72%	(An et al., 1996)
Remazol Black B (diazo)	Up-flow Filter	48h	>95%	(Oxspring et al., 1996)
Mordant Orange 1 (azo)	UASB	8h	95%	(Donlon et al., 1997)
Mordant Orange 1 (azo) Azodisalicylate (azo) Azodisalicylate (azo)	UASB	8h 8h 24h	99% 98.8% 88.9%	(Razo-Flores et al., 1997)
Maxilon red BL-N (basic)	Up-flow Filter	6-8.6h	99%	(Basibüyük et al., 1997)
Pricion Red H-E7B (azo)	UASB	16h	55-77%	(O'Neill et al., 2000)
Textile wastewater	Baffled	20h	90%	(Bell et al., 2000)
Textile wastewater	UASB	8-10h	80%	(Huren et al., 1994)
Textile wastewater	Fluidized bed	5.7h	59%	(Sen et al., 2003)

Under anaerobic conditions, azo dyes are degraded and converted into aromatic amines, which may be toxic, mutagenic and carcinogenic (Vanhulle et al., 2008). To achieve complete degradation of azo dyes, another stage involving aerobic biodegradation of the produced aromatic amines would be required (Zille, 2005). This anaerobic reduction implies decolorization as the azo dyes are converted to usually colorless but potentially harmful aromatic amines (Cervantes, 2009). Aromatic amines are normally not further degraded under anaerobic conditions, therefore the anaerobic treatment must be considered merely as the first stage of the complete degradation of azo dyes (Naresh et al., 2013). The second stage involves conversion of the produced aromatic amines under aerobic conditions.

4. Sequential Anaerobic-Aerobic Systems

This treatment system involves a spatially separated sequential exposure of dye wastewater to anaerobic conditions followed by an aerobic environment. Azo dyes are usually degraded under anaerobic conditions to colorless toxic amines which are readily metabolized under aerobic conditions (Ali, 2010). Complete treatment can thus be obtained by a sequenced anaerobic/aerobic treatment (Delèe et al., 1998). Many reactor studies have been performed on the treatment of textile wastewater, normally using both synthetic and actual dyestuff effluents (Oh et al., 2004). Several mono azo dyes, such as Mordant Yellow 5, Acid Red 18, Acid Red 14, and Acid Orange 10, have been successfully treated by a two-step anaerobic/aerobic process (FitzGerald et al., 1995; Haug et al., 1991). In another study, it was observed that Acid Yellow 17, Basic Blue 3 and Basic Red 2, treated in a UASB reactor followed by a semi-continuous activated sludge tank, were decolorized by 20, 72 and 78%, respectively, with no significant color removal in the aerobic stage (An et al., 1996). The color removal efficiency by the combination of an anaerobic baffled reactor and a fixed activated sludge reactor, treating Reactive Red 2, was 89.5% (Naimabadi et al., 2009). The performance of a system composed by a UASB reactor followed by a Submerged Aerated Biofilter (SAB) to remove color from actual textile effluent was around 52% in the UASB and 94% in the SAB (Amaral et al., 2014). Table 3 shows different combined systems and their color efficiency removal under anaerobic and aerobic conditions.

Table 3: Sequential anaerobic-aerobic reactor system (adapted from Van der Zee and Villaverde, 2005)

Anaerobic		**Aerobic**		**DYE**	**Color removal (%)**		**References**
Reactor	**HRT**	**Reactor**	**HRT**		**Anaerobic**	**Aerobic**	
UASB	24h	CAS	19h	Reactive Red 141	64%	11%	(O'Neill et al., 2000a)
UASB	4h	CAS	6.5h	Effluent from dye-manufacturing	70-80%	10-20%	(An et al., 1996)
UASB	25h	CAS	10h	Mordant Yellow 10	100%	0%	(Tan et al., 2000)
UASB	3-30h	CAS	10-30h	Reactive Black 5	82-98%	0%	(Sponza and Isik, 2002a)
UASB	2.6-26h	CAS	10-102h	Direct Red 28	97-100%	0%	(Sponza and Isik, 2005)
UASB	24h	SBR	24h	Acid Orange 7	60-97%	0%	(Ong et al., 2005)
Rotating disc	15h	Rotating disc	7.5h	Reactive Violet 5	90-95%	0%	(Sosath and Libra 1997)
Rotating disc	31h	Rotating disc	7.5h	Reactive Black 5	70%	0%	(Sosath et al., 1997)
Rotating disc	15h	Rotating disc	7.5h	Reactive Black 5	100%	35%	(Libra et al., 2004)
Filter	36h	CAS	36h	Acid Orange 10 Acid black 1 Direct Red 2 Direct Red 28	90-100% 100% 95-100% 80-100%	-	(Ranjaguru et al., 2000)
Filter	6h	Filter	7.7h	Acid Yellow 17 Basic Red 22	0-99%	-	(Basibuyuk and Forster 1997)
Filter	24h	CAS	24h	Actual Denim effluent	90%	-	(Setiadi et al., 2004)
Filter	12-72h	CAS	10	Reactive Red 195	60-100%	15%	(Kapdan et al., 2003)
Filter	12-72h	CAS	10	Actual Textile effluent	60-85%	10%	(Kapdan and Alparslan, 2005)

Analyzing the studies reported in Table 3, it can be concluded that long HRT are required in anaerobic wastewater treatment systems in order to achieve efficient decolorization of textile wastewaters. The decolorization efficiency achieved in the anaerobic step was usually higher than 70%, and in several cases almost 100%, whereas further decolorization was very limited in the subsequent aerobic stage. A wastewater treatment process in which anaerobic and aerobic conditions are combined is therefore the most logical concept for removing dyes from textile effluent (Frank and Santiago, 2005).

5. Integrated Anaerobic-Aerobic Systems

The basis of this systems lies in the fact that anaerobic and aerobic microorganisms can coexist beneficially in a single biofilm (Zitomer et al., 1998). Supplying oxygen to an oxygen-tolerant anaerobic consortium can create an integrated anaerobic-aerobic system (Tan et al., 1999). An integrated system called Expanded Granular Sludge Bed (EGSB) with an oxygenation of recycled effluent was carried out in order to remove Mordant Yellow 10 and 4-Phenylazophenol (Tan et al., 1999; Tan et al., 2001). In another study, the UASB reactor with an aerated upper part was used to remove Direct Yellow 26 (Kalyuzhnyi and Sklyar, 2000. A rotating annular drum, operating with different HRT to remove several acid dyes, showed color removal efficiencies ranging from 18% up to 90% (Harmer and Bishop, 1992; Jiang and Bishop, 1994). The baffled reactor with anaerobic and aerobic compartments was used to remove Reactive Red 5, showing high color removal efficiency (Gottlieb et al., 2002). Table 4 show different studies and results obtained using hybrid systems.

Table 4: Integrated anaerobic-aerobic reactor system (adapted from Van der Zee and Villaverde, 2005)

Integrated system	HRT	Dye	Color removal (%)		References
			Anaerobic	Aerobic	
EGSB with oxygenation of recycled effluent	36-43 h	Mordant Yellow 10	100%		(Tan et al., 1999)
	26-34 h	4-Phenylazophenol	< 100%		(Tan et al., 2001)
UASB with aerated upper part	1-100h	Direct Yellow 26	40-70%	10-20%	(Kalyuzhnyi and Sklyar, 2000)
Rotating Annular Drum	0.16-3h	Acid Orange 7	18-97%		(Harmer and Bishop, 1992)
Rotating Annular Drum	2h	Acid Orange 8 Acid Orange 10 Acid Red 14	20-90% 60% 60%		(Jiang and Bishop, 1994)
Baffled reactor with anaerobic and aerobic compartments	48/18 h	Reactive Black 5	84-88%		(Gottlieb et al., 2003)

The systems listed in table 4 are effective for the removal of a variety of dyes. The advantage of these systems is the small-scale requirement as compared to other sequential systems. An aspect to be considered is that these systems were performed mostly in lab-scale applications using synthetic dyes. A full-scale application should be assessed in order to evaluate the applicability of such a system in on industrial textile factory.

6. Factors Affecting Dye Biodegradation

Due to the highly variable nature of the biological treatment system and especially textile effluents, there are several factors that may affect the biodegradation rate of azo dyes. Non-dye related parameters such as temperature, pH and dissolved oxygen can all affect the biodegradation of azo dyes and textile effluents (Wuhrmann et al., 1980). Dye related parameters such as class and type of azo dye, reduction metabolites, dye concentration, dye side-group, and organic dye additives could also affect the biodegradability of azo dye wastewater (Seshadri and Bishop, 1994). Using activated sludge, a linear relationship between temperature and the reduction of Orange II was determined (Wuhrmann et al., 1980). The wastewater pH can affect the proper functioning of both anaerobic and aerobic organisms (Grady et al., 1999). In various studies, the production of inhibitory dye metabolites is cited as causing a decrease in biodegradation (O'Neill et al., 2000). Seshadri and Bishop (1994) performed a study investigating the effect of different influent dye concentration on the color removal efficiency. It was concluded that high dye concentrations may cause a drop in dye removal percent. Furthermore, the inhibition may be directly related to the effects of increased dye metabolite formation due to higher dye concentrations. It should be noted that tolerable influent concentrations are likely specific to individual or related groups of dyes (Cariell et al., 1995).

7. Conclusions

Due to the current strict regulations, dye decolorization and degradation is today one of the main issues in textile facilities. In order to design a proper effluent treatment plant, the aesthetic and environmental problems of discharging textile effluents in a water body should be considered. The activated sludge process is the most common treatment used all over the world in textile effluent treatment plants. As reported in literature, such a process is not able to completely remove color from the wastewater. Several applications, using only aerobic processes, are operating with long HRT to promote the bio-adsorption of dyes into the bio-sludge. This kind of process operation may ensure the effluent requirements in terms of local regulations for some countries, but it is not effective for dyes degradation, since they still persist in the sludge. Using white-rot fungi and or a specialized bacteria consortium, several dyes can be removed under aerobic conditions. In any case, the long growth phase of white-rot fungi represents an important disadvantage, because of the large reactor size that results due to fact that a long holding time for complete degradation, is required. Among the different biological treatments, anaerobic reduction, especially of azo dyes, can be an effective and economic treatment process for removing color from textile wastewater. The degradation of azo dyes, under anaerobic conditions, results in the production of aromatic amines. Post aerobic treatment is essential to oxidize the produced amines in the anaerobic stage. Generally, the most suitable plant layout involves spatially sequential anaerobic and aerobic treatments. The anaerobic reactor, e.g. UASB or baffled reactor, can be effective to remove around the 70% of azo dyes and part of the influent COD. The rest of COD is oxidized under aerobic conditions. Using the anaerobic reactor as pre-biological treatment, operating with an HRT of 8-12 h, it is possible to reduce the HRT and the energy required for aeration in the aerobic reactor. Another important advantage, is that by anaerobic degradation, the biogas produced can be used in a cogeneration system to produce heat and electricity. Is to be noted that anaerobic reactors need long start-up periods and, depending on the influent wastewater, additional external carbon sources.

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