Biological remediation of dyes in textile effluent: a review on current treatment technologies

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Abstract

Wastewater from the textile industry can contain a variety of polluting subst-ances including dyes. Increasingly, environmental legislation is being imposed to control the release of dyes, in particular azo-based compounds, into the enviro -nment. The ability of microorganisms to decolourise and metabolise dyes has long been known, and the use of bioremediation based technologies for treating textile wastewater has attracted interest. Within this review, we investigate the mechan-isms by which diverse categories of microorganisms, such as fungi, bacterial consortia and yeast, bring about the degradation of dyestuffs.

Keywords

Fungi, yeast, bacteria, anaerobic, biodgradation, aerobic, azo dye, biosorption, color

1. Introduction

Textile industries are found in most countries and their numbers have increased. These industries have shown a significant increase in the use of synthetic complex organic dyes as the colouring material. The annual world production of textiles is about 30 million tonnes requiring 700,000 tonnes of different dyes (Zollinger,19-87) which causes considerable environmental pollution pro-blems. Dyes include a broad spectrum of different chemical structures, primarily based on substituted aromatic and heterocyclic groups such as aromatic amine (C6 H5 -NH2), which is a suspected carcinogen, phenyl (C6 H5 -CH2) and naphthyle (NO2 -OH), the only thing in common is their ability to absorb light in the visible region. A large number of dyes are azo compounds (-N-N-), which are linked by an azo bridge.

Colour is the first contaminant to be recognized in wastewater and has to be removed before discharging into waterbodies or on land. The presence of very small amounts of dyes in water (less than1 ppm for some dyes) is highly visible and affects the aesthetic merit, water transparency and gas solu-bility in lakes, rivers and other waterbodies. The removal of colour from wastewaters is often more important than the removal of the soluble colourless organic substances, which usually contribute the major fraction of the biochemical oxygen demand(BOD). Methods for the removal of BOD from most effluents are fairly well established; dyes, however, are more difficult to treat because of their synthetic origin and mainly complex aromatic molecular structures. Government legislation is becoming more stringent in most developed countries regarding the removal of dyes from industrial effluents. Recently, state and federal agencies in the USA have been requiring lower effluent colour limits (< 200 units of American Dye Manufacturers Institute, ADMI) (McCurdy et al.,1992).

The wastewater characteristics from a dye house are highly variable from day to day, depending on the type of dye, the type of fabric and the concentration of the agents added. Treatment of such wastewaters is therefore, essential but di†cult. The discharge of dye house wastewater into

the environment is aesthetically displeasing, impedes light penetration, damages the quality of the receiving streams and may be toxic to treatment processes,to food chain organisms and to aquatic life. The degradation of molecules of dyes in the environment by microorganisms is likely to be slow (Meyer, 1981), which means that it is possible for high levels of dye to persist, and potentially accumulate. Furthermore, any degradation that does occur may produce smaller molecules equally unfamiliar to the environment, such as amines, and which may also be toxic.
There is no universal method for the removal of colour from dye waste (McCurdy et al.), the alternatives depend upon the type of dye wastewater. As the characteristics of dye wastewater are very variable, many different physical, chemical and biological treatment methods have been employed for its treatment.
The physical and chemical techniques were numerous including anion exchange resins(Karcher et al, 2002), flotation(Lin & Lin, 1993), electroflotation(Ogfitveren & Koparal, 1994), electrochemical destruction(Ulker & Savas, 1994), irradiation (Shen et al, 2002), ozonation(Zhang et al, 2003), adsorption(El-Gundi, 1991)and the use of activated carbon (Pala et al, 2002), etc. Some of physical and chemical treatment techniques are effective for colour removal but use more energy and chemicals than biological processes. They also concentrate the pollution into solid or liquid sidestreams requiring additional treatment or disposal. In recent years, a number of studies have focused on some microorganism which are to biodegrade and biosorb dye in wastewaters. Awide variety of micro -organisms capable of declorizing a wide range of dyes include some bacteria, fungi, yeast.
This paper summarizes microbical declorization of dye used in textile industries, reports on progress and discusses mechanisms and the factors affecting the process.

2. Microbial decolorization

2.1 Biodegradation

2.1.1 Fungal biodegradition

White-rot fungi are those organisms that are able to degrade lignin, the structural polymer found in woody plants (Barr and Aust, 1994). The most widely studied white-rot fungus, in regards to xenobiotic degradation, is *Phanerochaete chrysosporium*. This fungus is capable of degrading dioxins, polychlorinated biphenyls (PCBs) and other chloro-organics (Reddy, 1995). In addition to *P. chrysosporium*, other white-rot fungi, also capable of decolorizing dyes, include *Coriolus versicolor*(Sam et al, 2001*),Trametes versicolor* (Wong and Yu, 1999; Libra et al 2003), *Coriolus versicolor* (Knapp and Newby, 1999) and *Funalia trogii* (Yesilada et al., 1995). Meanwhile, there are various fungi other than white-rot fungi, such as *umbelopsis isabellina* and *penicillium geastrivous*(Yang et al, 2003), aspergillus foetidus, *Rhizopus oryzae* (Polman and Breckenridge, 1996) which can also decolorize and/or biosorb diverse dyes. In fungal decolorization of dye wastewater, these fungi can be classified into two kinds according to their life state: living cells to biodegrade and biosorb dyes and dead cells (fungal biomass) to adsorb dyes.
For living cells, the major mechanism is biodegradation because they can produce the lignin modifying enzymes, laccase, manganese peroxidase (MnP) and lignin peroxidase (LiP) to mineralize synthetic lignin or dyes (Raghukumar et al., 1996; Fu and Viraraghavan, 2001). However, the relative contributions of LiP, MnP and laccase to the decolorization of dyes may

be different for each fungus. For the fungus *P. chrysosporium*, LiP was found to be responsible for the decolorization of the dyes (Pasti-Grigsby et al., 1992). Ollikka et al. (1993) reported that LiP played a major role in the decolorization of azo, triphenyl methane, heterocyclic and polymeric dyes by *P. chrysosporium* and that MnP was not required to start the degradation of these dyes. Young and Yu (1997) studied the decolorization of the eight synthetic dyes, including azo, anthraquinone, metal complex and indigo, by *T. versicolor* and also reported that MnP did not decolorize these dyes while ligninase-catalyzed oxidation removed over 80% of the dyes. However, Zhang et al. (1999) observed that MnP played an important role in the decolorization of cotton bleaching effluent by an unidentified white-rot fungus, while there was no obvious role for LiP in this decolorization. Because *T. versicolor* releases laccase as its major extracellular enzyme, its major mechanism in decolorizing anthraquinone, azo and indigo dyes was laccase activity (Wong and Yu, 1999).

Fungal decolorization is a promising alternative to replace or supplement present treatment processes. However, using fungal biomass to remove color in a dye wastewater is still in the research stage. More studies are needed to develop a practical application.

2.1.2 Bacteria biodegradation

Efforts to isolate bacterial cultures capable of degrading azo dyes started in the 1970s with reports of a *bacillus subtilis* (Horitsu et al., 1977), followed by numerous bacteria: *pseudomonas spp* were isolated from an anaerobic-aerobic dyeing house wastewater treatment facility as the most active azo-dye degraders(Yu et al, 2001); Chang et al(2004) used the extracellular metabolites of a dye-decolorizing strain, *Escherichia coli strain NO3*, as a biostimulator to entice E. coli strain NO3 into a beneficial mode of metabolism for an economically feasible decolorization.To design technical decolorization processes of textile wastewater treatment with sulfide produced by *Sulfate reducing bacteria (SRB*) was designed to decolorize textile wastewater treatment with sulfide(Yoo, 2002)

In general, the wastewater from textile industry contains many various dyes. To gain a widespread reception, the azo-degrading bacteria should exhibit decolorizing ability for a wide range of dyes, *aeromonas hydrophila* was selecte from six bacterial strains with the capability of degrading textile dyes (Chen et al, 2003). Isolating such microorganisms proved to be a difficult task. Mixed bacterial cultures from a wide variety of habitats have also been shown to decolourise the diazolinked chromophore of dye molecules in 15 days (Knapp and Newby, 1995). Nigam and Marchant (1995) and Nigam et al. (1996) demonstrated that a mixture of dyes were decolourised by anaerobic bacteria in 24-30 h, using free growing cells or in the form of biofilms on various support materials.

The ability of bacteria to metabolise azo dyes has been investigated by a number of research groups. Under aerobic conditions azo dyes are not readily metabolised, the intermediates formed by these degradative steps resulted in disruption of metabolic pathways and the dyes were not actually mineralised. Under anaerobic conditions, such as anoxic sediments, many bacteria gratuitously reduce azo dyes reportedly by the activity of unspecific, soluble, cytoplasmic reductases, known as azo reductases. These enzymes are reported to result in the production of colorless aromaticamines which may be toxic, mutagenic, and possibly carcinogenic to animals (Mcmulan et al, 2001).

2.1.3 Yeast biodegradation

Only a limited amount of studies about yeast decolorization were reported. The ability of *Kluyveromyces marxianus IMB3* to decolorize Remazol Black-B dye was investigated and maximum colour removal, 98%, was achieved at 37 degrees C(Meehan et al, 2000). Zissi et al. (1997) showed that *Bacillus subtilis* could be used to break down azo dye. A number of simple azo dyes was degraded in liquid aerated batch cultures by a strain of the yeast Candida zeylanoides, the extent of colour removal ranged from 44 to 90%, after 7 days (Martins et al. 1999)

2.2 Microbial biosorption

The uptake or accumulation of chemicals by microbial mass has been termed biosorption (Hu, 199 and Kumar et al., 1998). Bacteria and fungi have all been used for the purpose of decolourising dye-containing effluents. Knapp et al. (1995) reported that the extent of color removal by adsorption was always limited, generally less than 50%. Benito et al. (1997) studied color adsorption by *T. versicolor* mycelium and reported that the adsorption accounted for only 5–10% of the total color removal. Miranda et al. (1996) observed that the percentage of adsorbed color was in the range 10–25 in the study with *Aspergillus niger*. Hu (1992) demonstrated the ability of bacterial cells to adsorb reactive dyes. Zhou and Zimmerman (1993) used actinomyces as an adsorbent for decolourisation of effluents containing anthroquinone, phalocyanine and azo dyes. The decolorization of dyes with different molecular structures by *Cunninghamella elegans* was evaluated under several media condition(Ambrosio et al, 2004). Three reactive dyes were rapidly adsorbed by the mycelium pellets of Penicillium oxalicum. Dye removal of Reactive Blue 19 was up to 60% in 10 min and 91% in 80 min(Zhang et al 2003). Depending on the dye and the species of micro-organism used different binding rates and capacities were observed. It can be said that certain dyes have a particular affinity for binding with microbial species. The use of biomass has its advantages, especially if the dye-containing effluent is very toxic. Biomass adsorption is effective when conditions are not always favourable for the growth and maintenance of the microbial population. Adsorption by biomass occurs by ion exchange.

2.3 Proccess influence factors

In the decolorization process by fungi, bacteria and yeast, there are various influencing factors. They can be grouped into two kinds: one is related to microbial growth conditions; the other is related to the characteristics of the dye solution or wastewater.

2.3.1 Microbial growth contitions

As different components possess different abilities to decolorize dyes, it is necessary to create an optimal environment favorable to microbial growth and thus make the microbial possess the maximum ability to decolorize dyes in wastewater. Important microbial growth conditions are discussed as follows:
Medium, fungi are mostly grown in a medium with dyes or dye wastewater to develop a biosorbent containing living fungi. The medium is mainly composed of carbon source, nitrogen source and other nutrients;Carbon, all kinds of carbon resource were investigated how they effect the decolorization.(Zhang et al. 1999; Belsare and Prasad 1988; Mou et al., 1991);Oxygen,Soares and Duran (1998) reported that agitation was essential for keeping a high rate of decolorization

by *T. villosa*; Nitrogen(Zhen and Yu, 1998), Nutrient concentrations, incubation time(Sumathi and Phatak, 1999), pH and tempreture were also studied as parameters, mycelium pellets obtained the maximum adsorption(zhang et al, 2003)at pH 2.

2.3.2 Characteristics of dye wastewater

Dyes, Different dyes have different molecular structures. So a microbial capable of decolorizing one dye may have different capacities for other dyes(Wong and Yu, 1999; Knapp et al. 1995; and Paszcczynski et al. 1992).
Dyeing processes consume large amounts of salt, so the concentrations of salt in dye wastewaters are normally high. Mou et al. (1991) studied samples from a textile dyeing factory wastewater with as high as 15% (w/v) chloride ion. So ionic strength is an important factor. Zhou(1991)reported that high ionic strength (the concentration of NaCl) led to high biosorption of humic acid by *R. arrhizus*. The effect of ionic strength was similar to that of a colloid. At higher ionic strength, the electrical double layers of both *R. arrhizus* biomass and humic acid would be compressed thinner. Therefore biomass and humic acid could approach closer and thus this would increase van der Waals bonding and hence increase biosorption. Tempreture, as various textile and other dye effluents are produced at relatively high temperatures (50–60°C) (Banat et al., 1996), so temperature will be an important factor in the real application of biosorption by biomass in future.
There are various factors influencing microbial decolorization related to microbial growth and the characteristics of dye wastewater. Dye molecules have many different and complicated structures. This is one of the most important factors influencing microbial decolorization. Further research is needed to establish the relationships between dye molecule structure and microbial decolorization.

3. Textile-dye bioremediation system

3.1 Aerobic system

The recalcitrant nature of zero dyes, together with their toxicity to mircrooganism, makes aerobic treatment difficult.Therefore,especially for color removal,various adsorbents and chemicals have been directly added into the activated sludge systems in certain studies.In such a study, (Marquez and Costa obtained 90%acid orange 7 dye removal effciency was obtained by addition of activated carbon into the aeration tank (Marquez and Costa, 1996).It was also determined that the increase in activated carbon particle size decreases color removal effciency .In another study, the addition of activated carbon was investigated and color removaleffciencies for different kinds of dyes were determined (Marmagne and Costa 1996). A speci fc organic focculant (Marwichem DEC), powdered activated carbon (PAC),bentonite,activated clay and commercial synthetic inorganic clay (Macrosorb)were directly added into the activated sludge laboratory pilot plant model.(Pala, et al, 2002).

3.2 Anaerobic system

Under anaerobic conditions, azo dyes are readily cleaved via a four-electron reduction at the

azo linkage generating aromatic amines. The required electrons are provided by electron donating carbon source such as starch, volatile fatty acids (VFA) or glouse. In addition , it is known that methanogenic and acetogenic bacteria in anaerobic microbial consortium contain unique reduced enzyme cofactors that could also potentially reduce azo bonds(Carliell et al, 1996).

Among the many anaerobic systems available are:

Baffled reactor	Bell et al (2000)
UASB	Chinwekitvanich et al (2000)
Anaerobic filter reactor	Basibuyuk (1997)
UAF	Oxspring (1996)
FBR	Sen et al (2003)

3.3 Aerobic/anaerobic system

Dye colour is sensitive to redox and anaerobic treatments are effective at decolourising reactive azo dyes (Beydilli, et al). Azo dyes account for about two-thirds of the dye catalogue Anaerobic cleavage of the azo bond (---N=N---) results in permanent decolourisation of the dye, but the intermediates can be reoxidised to coloured by-products. Specific azo reductase enzymes are also reported.

The aromatic amine residues from anaerobic decolourisation resist further anaerobic degradation and they are also reported mutagens, but they can be mineralised aerobically . Many researches reports on the advantages of cocultures of anaerobic and aerobic organisms in sequential redox environments for the treatment of refractory COD. A great number of the reported examples of azo dye biodegradation comprise two main steps, the reductive cleavage of the azo bond under anaerobic conditions and the subsequent aerobic mineralization of the produced aromatic amines.

UASB+UBAF	Lacalle et al (2001)
SBR	Shaw et al (2002)
Anoxic + Anaerobic/Aerobic SBR	Panswad et al (2001)

Aerobic-anaerobic packed-bed reactors	Lin & Liu, (1994).
Aerobic-anaerobic fluidized-bed reactors	Seshadri et al., (1994).
Aerobic-anaerobic sequential batch or continuous-flow reactors	Oxspring et al., (1996).

4. CONCLUSION

Coloured-dye-wastewater treatment and decolorization presents an arduous task. Wide ranges of pH, salt concentrations and chemical structures often add to the complication. Among the most economically viable choices available for effluent treatment/decolorization, and the most practical in terms of manpower requirements and running expenses to adopt and develop, appear to be the biological systems. Fungal , bacteria and yeast decolorizations are a promising alternative to replace or supplement present treatment processes. The techniques by which decolorization occurs vary and among them adsorption seems of great significance for future development in bio-removal or bio-recovery of dye. However, using microbial biomass to remove color in a dye wastewater is still in the research stage. More concerted efforts are still required to establish biological decolorization systems.

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