Available online at www.ijpab.com

DOI: http://dx.doi.org/10.18782/2320-7051.2400

ISSN: 2320 – 7051

Int. J. Pure App. Biosci. **4 (5):** 81-89 (2016)

International Journal of Pure & Applied Bioscience

Review Article

OPEN ACCESS

Degradation of Toxic Dyes- A Review

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Received: 18.10.2016 | Revised: 27.10.2016 | Accepted: 29.10.2016

ABSTRACT

Nowadays globalization, urbanization and industrialization leads to various environmental concerns. The usage of synthetic dyes increases in many areas. Among the synthetic dyes, reactive dyes are most commonly used in all industries. Dye is an integral part which is used to impart colour to materials. The waste generated during the process and operation of the dyes, contains the inorganic and organic contaminant leading to the hazard to ecosystem and biodiversity causing impact on the environment. The physico-chemical treatment does not remove the color and dye compound concentration. The decolorization of the dye takes place either by adsorption on the microbial biomass or and enzymatic degradation. Bioremediation takes place by anaerobic and/or aerobic process. In the present review the decolorization and degradation of dyes by fungi, algae, yeast and bacteria have been cited. The factors affecting decolorization and biodegradation of dye compounds such as pH, temperature, dye concentration, effects of carbon dioxide and nitrogen, agitation, effect of dye structure, electron donor and enzymes involved in microbial decolorization of dyes have been also high lightened in the review.

Key words: *Decolorization, Physico-Chemical, Dye, Ecosystem*

INTRODUCTION

Environmental pollution due to urbanization and rapid growth of industries has a detrimental effect on human health and ecology. Textile dyes constitute a major source of pollution. Textile industries consume a major share of dyes in India. Further, the textile industry of India also contributes nearly 14% of the total industrial production of the country. Various chemical substances discharged from the industries become a persistent environmental contaminant. Due to rapid industrialization and urbanization, a lot of chemicals including dyes, pigments, and aromatic molecular structural compounds were widely used in several industrial applications such as textiles, printing, pharmaceuticals, food, toys, paper, plastic and cosmetics[36]. These industries have shown a significant increase in the use of synthetic dyes as a coloring material. The annual world production of textiles is about 30 million tones requiring 700,000 tonnes of different dyes[56].

Cite this article: Kalyani, P., Hemalatha, V., Vineela, K. and Hemalatha, K.P.J., Degradation of Toxic Dyes- A Review, *Int. J. Pure App. Biosci.* **4(5):** 81-89 (2016). doi: http://dx.doi.org/10.18782/2320-7051.2400

The dyes includes such as acidic, reactive, basic, disperse, azo, diazo, anthraquinone dyes which causes a considerable environmental pollution problems. Many dyes and pigments are hazardous and toxic for human as well as aquatic life at the concentration at which they are being discharged to receiving water. The high concentration of dyes is known to cause ulceration of skin, and mucous membrane, dermatitis, perforation of nasal septum, severe irritation of respiratory tract and on ingestion may cause vomitting, pain, haemorrhage and sharp diarrhea[30]. Over the last decades, the increasing demand for dyes by the textile industry has shown a high pollutant potential. It is estimated that around 10 -15% of the dyes are lost in the waste water during the dyeing processes.

There are various types of dyes which are discussed as follows:

Azo dyes

Azo dye is the largest group of dyes, with -N=N- as a chromophore in an aromatic system. There are monazo, disazo, trisazo, tetrakisazo and polyazo dyes depending upon the number of azo-groups present. Diazotisation of a primary amine, in presence of HCl + NaNo2 at freezing temperature, produces a diazonium salt which in turn coupled with aromatic compounds, producing an azo-dye.

Anthraquinone dyes

Anthraquinone is the basic unit of this class of dyes. It is faint yellow in color which is sufficient to use it as a dye but it cannot be classified as a dye. Dyes containing anthraquinone unit belong to mordant, disperse and vat dyes. Its quinonoid system acts as a chromophore. Anthraquinone dyes have excellent fastness properties.

Disperse dyes

Disperse dyes generally use to dye cellulose acetate, nylon and other hydrophobic fibres. They are also known as acetate dyes. Sulphoricin oleic acid (SAR) is used as the dispersing agent. Dispersal and cellitoin are the important dispersing agents. Tatrazine (otherwise known as E number E102 or C.I. 19140) is a synthetic lemon yellow azo dye used as a food coloring.

Adsorption by dead microbial cells

Killed bacteria, yeast and fungi are used for the decolorization of dye containing effluents. The dyes from textile industries are varied from their chemistries and so their interactions with microorganisms depend on the chemistry of particular dye and microbial biomass[37]. Adsorption method is used during unfavourable conditions for the growth and preserves microbial population[35]. Adsorption process by microorganism is carried out by ion exchange method. Bacterial cells adsorb reactive dyes[16]. The use of dead organisms instead live biomass overcomes the problem such as waste toxicity and requirement of nutrients.

Biodegradation of dyes Fungal Biodegradation

A group of fungal organisms have an ability to decolorize wide range of dyes[12,13]. Fungus is capable of degrading dioxins, polychlorinated, biphenyls (PCBS) and chloro- organics[41]. The decolorization was a secondary metabolic activity linked to the fungus ligninolytic degradation activity. The degradation of some xenobiotic by other white- rot fungi is known to occur under non- ligninolytic conditions and would mainly be through the laccase enzyme activity[10]. Fungi, due to their excretion of extracellular enzymes, are known to be able to degrade though possibly not completely the structures that are difficult for bacteria to handle. Microbial degradation of Congo red by *Gliocladium virens*[45], various hazardous dyes likes, Congo red, Acid red, Basic blue and Bromophenol blue, Direct green by the fungus *Trichoderma harzianum* The results were similar to biodegradation of Congo red and Bromophenol blue by the fungus *Trichoderma harzianam* in semi-solid medium and biodegradation of Methylene blue, Gentian violet, Crystal violet, Cotton blue, Sudan black, Malachite green and Methyl red by few species of *Aspergillus* in liquid medium. Cripps *et al*[9]., also reported the biodegradation of three azo dyes (Congo red, Orange II and Tropaeolin O) by the fungus *Phaenerocheate chrysosporium*. Rahna K. Rathnan *et al*[39]., found that the isolated fungus *Aspergillus* *niger* and *Aspergillus oryzae* and mixed consortium is as an important source for bioremediation of toxic dye. *Aspergillus niger* showed greater decolorisation production during sixteen days incubation[32].

Bacterial degradation

Work on bacterial degradation of dyes were started in the 1970s with report on *Bacillus* s*ubtilis*[15], then degradation was followed by numerous bacteria such as *Aeromonas hydrophilia*[17], *Bacillus cereus*[53], *Pseudomonas sp*[29], *E. Coli*[4]. Whereas in anaerobic conditions, bacteria reduce azo dyes gratuitously by the activity of unspecific, soluble, cytoplasmic reductase called azo reductase. Halophiles have been reported to be involved in the dye decolourization. The moderately halotolerant *Bacillus sp*. were isolated for decolourization of azo dye Red 2G to an extent of 64.89%.

Algal Biodegradation

Algal culture also has an ability to degrade azo dyes through azoreductase[20]. *Chlorella* and *Oscillatoria* were capable of degrading azo dyes to aromatic amines and further to simple organic compounds. *Synechocystis* sp and *Phormidium* sp have a capacity to remove reactive dyes such as Reactive Red, Remazol Blue, and Reactive Black B[24].

Yeast Biodegradation

Limited amount of studies about yeast decolorization were reported. *Kluyveromyces marxianus* IMBS decolorize Remazol Back B dye of about 98%[34]. *Pseudozymarugulosa* Y - 48 and *Candida krusei* G-1 are the yeast strains exhibited excellent color removal of reactive azo dyes. *Saccharomyces cerevisiae* MTCC 463

Table 1: Factors affecting decolorization and degradation of synthetic dyes which has been shown below

Factors	Descriptions
pH	The pH has a major effect on the ef ciency of dye decolorization, the optimal pH for color removal in bacteria is often between 6.0 and 10.0. The tolerance to high pH is important in particular for industrial processes using reactive azo dyes, which are usually performed under alkaline conditions.
Temperature	Temperature is also again a very important factor for all processes associated with microbial vitality, including the remediation of water and soil. It was also observed that the decolorization rate of azo dyes increases uptothe optimal temperature, and afterwards there is a marginal reduction in the decolorization activity.
Dye concentration	Earlier reports show that increasing the dye concentration gradually decreases the decolorization rate, probably due to the toxic effect of dyes with regard to the individual bacteria and/or inadequate biomass concentration, as well as blockage of active sites of azo reductase by dye molecules with different structures.
Carbon and nitrogen Sources	Dyes are de cient in carbon and nitrogen sources, and the biodegradation of dyes without any supplement of these sources is very diffcult. Microbial cultures generally require complex organic sources, such as yeast extract, peptone, or a combination of complex organic sources and carbohydrates for dye decolorization and degradation.
Oxygen and Agitation	Environmental conditions can affect the azo dyes degradation and decolorization process directly, depending on the reductive or oxidative status of the environment, and indirectly, in uencing thenmicrobial metabolism. It is assumed that under anaerobic conditions reductive enzyme activities are higher; however a small amount of oxygen is also required for the oxidative enzymes which are involved in the degradation of azo dyes.
Dye structure	Dyes with simpler structures and low molecular weights exhibit higher rates of color removal, whereas the removal rate is lower in the case of dyes with substitution of electron withdrawing groups suchas SO3H, SO2NH2 in the para position of phenyl ring, relative to the azo bond and high molecular weight dyes.
Electron Donor	It has been observed that the addition of electron donors, such as glucose or acetate ions, apparently induces the reductive cleavage of azo bonds. The type and availability of electron donors are important in achieving good colour removal in bioreactors operated under anaerobic conditions.
Redox Mediator	Redox mediators (RM) can enhance many reductive processes under anaerobic conditions, including azo dye reduction.

(*Source: Khan et al., 2013)*[26]

Table 2: Advantages and disadvantages of the dye removal methods

Physical/chemical methods	Advantages	Disadvantages
Fentons reagent	Effective decolourisation of both soluble and insoluble dyes	Sludge generation
Ozonation	Applied in gaseous state: no alteration of volume	Short half-life (20 min)
Photochemical	No sludge production	Formation of by-products
Cucurbituril	Good sorption capacity for various dyes	High cost
Activated carbon	Good removal of wide variety of dyes	Very expensive
Electrochemical Destruction	Breakdown compounds are non-hazardous	High cost of electricity
Wood chips	Good sorption capacity for acid dyes	Requires long retention times
Silica gel	Effective for basic dye removal	Side reactions prevent commercial application
Membrane filtration	Removes all dye types	Concentrated sludge production
Ion exchange	Regeneration: no adsorbent loss	Not effective for all dyes
Irradiation	Effective oxidation at lab scale	Requires a lot of dissolved O_2
Electrokinetic coagulation	Economically feasible	High sludge production

(*Source : Joshni. T.C. et al., 2011)*[21]

REMOVAL TECHNIQUES

Several physical, biological and chemical removal techniques like adsorption, coagulation, flocculation, membrane filtration, ozonation, electrochemical, radiolysis, bacterial, algal, fungal and advanced oxidation processes have been known to decolorize the textile effluents[6,18,23,40,46,51]. Physico-Chemical treatments on one hand transfer pollutants present in the effluents from one phase to other without eliminating them[11], the Biological methods can remove a wide range of colors by aerobic/anaerobic bacterial and fungal degradation[1]. Advanced Oxidation Processes (AOP) deal with the generation and use of reactive free radicals to oxidize most of the complex chemicals present in the effluents. AOPs can convert the complex dissolved organic pollutants to simpler and non-toxic degraded products. The generation of highly reactive free radicals can be attained by using UV, UV/O_3, UV/H_2O_2, Fe+2/H_2O_2, TiO_2/H_2O_2 and others[25].

PHYSICO-CHEMICAL METHODS

Various Physico-Chemical techniques are known which can effectively decolorize textile wastewater, but amongst them Adsorption[8] is one of the removal techniques which has gained utmost attention mainly because of its simplicity and insensitivity to toxic pollutants. Although this technique produces high quality of treated water but the problem lies in selection of most appropriate adsorbent. Several adsorbents have been explored till date for textile water decolorization. In most cases adsorption is accompanied by sorption process. This combination of adsorption and sorption is termed as biosorption.

Biological Methods

Biological treatment methods are eco-friendly methods which are gaining importance in today's scenario. Microorganisms such as bacteria, fungi, algae, yeast and enzymes can be successfully utilized to remove color of a wide range of dyes through anaerobic, aerobic, and sequential anaerobic-aerobic treatment processes.

Bacterial Methods

The evaluation of *Shewanella* sp. strain KMK6 as adsorbent for the decolorization of mixture of textile dyes. This bacterial strain was isolated from the dye contaminated soil and was applied to mixture of dyes under suitable conditions. The results indicated a decrease in the COD (Chemical Oxygen Demand) and color of the dye mixture with the production of nontoxic degraded products[28]. Some other strains of bacteria like *Pseudomonas fluorescens* strains (Sz6 and SDz3)[14] and *Shewanella* bacterial strains[31] have also been used successfully. Decolorization of mixture of dyes and actual textile effluent was done with a novel bacterium *Lysinibacillus* sp. RGS in another study about 87% decolorization was obtained for mixture of dyes with 69% COD reduction after 48 hours[44]. In 2013 a plant bacterial synergistic system for efûcient treatment of the textile efûuents was developed. *Glandulariapulchella* (Sweet) *Tronc*., *Pseudomonas monteilii* ANK and their consortium were used to decolorize the dye mixture. Consoritum showed 100% decolorization for mixture of dyes[22].

Fungal Methods

A thermophilic fungus, *Thermomucorindicae seudaticae* obtained from compost was successfully used for azo anthraquinone dye mixture, the optimum temperature and pH for adsorption was found to be 55°C[48]. Decolorization of mixture of two dyes i.e. brilliant green and evans blue by fungi was studied. Individual and mixture of fungal strains Pleurotuso *streatus* (BWPH), *Gloeophyllum odoratum* (DCa), and *Fusarium oxysporum* (G1) were used during the fungal degradation[38].

Biosorption of mixture of three reactive azo dyes (red, black and orange II) by inactive mycelium of *Cunning hamellaelegans* was investigated. The presence of heterogenous binding sites was suggested by Freundlich adsorption isotherm model which fitted best to the experimental data[3], a mixture of structurally different azo and non-azo dyes was degraded using *Galactomyces geotrichum* MTCC 1360 which is a species of yeast. Approximately 88% of ADMI removal of mixture of structurally different dyes was observed within 24 h at 30°C and pH 7.0 under shaking condition (120 rpm). The reduction of COD (69%), TOC (43%), and phytotoxicity study indicated the conversion of complex dye molecules into simpler oxidizable products having less toxic nature[49].

Decolorization of real textile effluent and synthetic dye mixture by *Trametes versicolor*, a mixture of dyes containing each dye in equal amounts was taken as the synthetic wastewater. A decolorization of 97% was achieved for initial dye concentrations up to 100 mg/l. pH and the presence of glucose were identified as important parameters for an adequate decolorization performance. In addition to this, comparative studies were also done using several fungi (*Phanerochaete chrysosporium, Pleurotusostreatus, Trametes versicolor* and *Aureo- basidiumpullulans*) under optimized conditions amongst which *T. versicolor* showed the best biodegradation performance[2]. A group of researchers evaluated the possibility of a fungal wastewater treatment for a mixture of bioaccessible reactive azo dyes using biodegradation assays[33]. Whiterot fungus *Ph. tremellosa* was found capable of decolorizing an array of synthetic textile dyes[27]. Isolated fungi, *Aspergillus foetidus as adsorbent* which effectively decolorized reactive Diamerene textile dyes was also evaluated, the fungus was able to decolorize a mixture of dyes upto 85% within 72 hours of its growth in presence of 5 ppm of chromium and 1% sodium chloride.

Enzymatic Methods

Ammonium sulphate fractionated pointed gourd (Tricho santhesdioica) peroxidase-concanavalin A (PGP-Con A) complex, entrapped into calcium alginatepectin gel was used for the decolorization of a mixture of two dyes, the experiment was carried out in a batch and continuous two reactor catalytic systems for the removal of synthetic dyes[19]. A decolorization of mixture of azo and anthraquinone dyes using Trametestrogii laccase. During the treatment anthraquinone dyes played the role of mediator and assisted in degradation of azo dyes with purified enzyme[55].

Evaluating the ability of Cyathusbulleri laccase to decolorize and detoxify the mixture of reactive and acidic dyes in presence of natural and synthetic mediators, the laccase–ABTS system did 80% decolorization of the simulated dye mixture[7]. The use of plant polyphenol oxidases to degrade a complex mixture of dyes from textile waste water, potato polyphenol oxidases and brinjal polyphenol oxidases were used in enzymatic degradation. Potato plant polyphenol oxidase results were more eûective in decolorizing the dye mixtures.

ADVANCED OXIDATION PROCESSES

These processes include techniques like Fenton's reagent oxidation, ultra violet (UV) photolysis and sonolysis, and are capable of degrading the organic pollutants at ambient temperature and pressure. AOPs have been widely used for the decolorization of textile dye effluent and also for removal of recalcitrant organic components present in it. The versatility of AOP lies in the fact of different possible ways for OH• radicals generation. AOPs show explicit advantages over conventional treatment methods as they can eliminate non-biodegradable organic components and there is no problem of residual sludge disposal[25].

Ozonation

Ozonation treatment was examined for the removal of reactive dyes from textile dyeing industrial effluent in a batch reactor at 35ºC. Effects of pH and reaction time on the decolorization efficiency were also evaluated, with time the color intensity of the waste water reduced. The decoloration efficiency increased from 32.83 % to 56.82 % as the time progresses after six hours about more than 90% of the color was removed[50]. Synthetic dye effluent was prepared using nine commercially reactive dyes and similar ozonation was carried out[42]. Ozonation can be used as a viable technique for treatment of colored effluents. Ozonation was done in a semi-batch reactor was carried out for a mixture of eight reactive dyes in which concentration of dyes in mixture ranged from 50-500 mg/l. Maximum color and COD removal was achieved at an ozone dose of 4.33 mg/l at 30 mins. Initially at lower dye concentration, decolorization and COD removal rate was fast, but as the concentration increased from 200- 500 mg/l it took longer time to decolorize. During Ozonation biodegradability increased following pseudo first order kinetics.

Electrochemical Oxidation

In this process electrochemical oxidation of the actual textile effluent and synthetic dye solution by using titanium-tantalum-platinum-iridium anode, batch experiments were conducted by taking a synthetic mixture of sixteen dyes having 361 mg/L concentration with 281 mg/L of COD, whereas actual effluent having residual dyes, by products and COD 404 mg/L. Quantitative decolorization was obtained within 10-15 min. The method consumed low energy and the extent of mineralization was 30-90% at 180 mins showing moderate degree of mineralization. Performance increased with increase in current intensity, salinity and with decrease in pH. Eco toxicity was assessed showing presence of toxic by-products[5]. In mixtures of two dyes, the decolorization rate became similar for all the dyes. The results revealed that electrochemical oxidation method was suitable for effective decolorization of wastewater from industries[43].

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