Review Paper

Decolorization of mixture of dyes: A critical review

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ABSTRACT: Water plays a vital and essential role in our ecosystem. This natural resource is becoming scarce, making its availability a major social and economic concern. Use of a large variety of synthetic dyes in textile industries has raised an hazardous environmental alert. About 17 - 20% of freshwater pollution is caused by textile effluents. These effluents are recalcitrant to biodegradation and cause acute toxicity to the receiving water bodies, as these comprised of various types of toxic dyes, which are difficult to remove. Decolorisation of textile wastewater is therefore important before releasing it into the nearby local waterways. It therefore becomes essential to degrade the toxic chemicals of textile wastewater, so as to avoid the hazardous environmental effects. Several treatment methods have been employed to embark upon the problem of dye removal but degradation becomes further more difficult for effluents containing dye matrix. The review study has been an attempt to present the different diversified attempts used for decolorisation of a mixture of dyes.

Keywords: *Decolorisation; Dye mixture; Synthetic dye; Textile waste water; Ecosystem*

INTRODUCTION

Water is vital requirement of life and used for various household as well as industrial activities. It is one of the most essential natural resources, unfortunately exploited the most. The key reason being increased human population, rapid industrialization, increased living standard and urbanization. Today the whole world is facing water crises because of unrestricted and excessive exploitation of water. Rapid urbanization of natural resources like, increase in industries, especially textile industries is posing a threat to the water bodies as these discharge effluents with various harmful and toxic components, mainly dyes. This deteriorates the quality as well a quantity of water and makes it unsafe for further use.

Synthetic dyes are of great environmental concern due to their widespread usage and their low removal rate during aerobic waste treatment. About 10,000

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different dyes are prepared globally and approximately 8×10^5 tons of synthetic dyes are consumed in textile industries in the whole world (Walker and Weatherley, 1997). Textile wastewater is a complex and highly variable mixture of many polluting substances, including dyes, which induce color coupled with organic load leading to disruption of the total ecological balance of the receiving water system (Hassani *et al.*, 2008). In Textile industries 93% of the intake water comes out as colored wastewater due to dyes containing high concentration of organic compounds and heavy metals (Wijannarong *et al.,* 2013; Gupta *et al.*, 2014). The non-biodegradable nature of the dyes in the spent dye baths of textile industries constitutes a serious environmental hazard. The color of wastewater is aesthetically unpleasant to aquatic bodies which hinder the oxygenation ability of water, disturbing the whole of the aquatic ecosystem (Xu *et al.*, 2005) and food chain. Textile effluents comprise of different dyes hence it

becomes essential to promote the prevailing techniques as well as to look for new techniques that decolorize the mixture of dyes rather than a single dye solution.

The paper presents an in-depth study of the different prevailing techniques used till date for the decolorisation of the matrix of dyes. Various removal techniques used for decolorization have been discussed followed by the results obtained in different researches. The paper would acquaint the readers to the different removal methods employed for a similar reason, proving to be a knowledge pool and helping the researchers working in the similar field to design appropriate treatment plans.

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 (Kannan and Sundaram, 2001; Rai *et al.*, 2005; Solmaz *et al.*, 2009; Wojnarovits and Takacs, 2008; Ince and Tezcanh, 1999; Chaudhari *et al.*, 2011).

Physico-Chemical treatments on one hand transfer pollutants present in the effluents from one phase to other without eliminating them (Erswell *et al.,* 1988), the Biological methods can remove a wide range of colors by aerobic/anaerobic bacterial and fungal degradation (Aksu, 2005). 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 (Kestioglu *et al.*, 2005). A summary of the some researches related to the above mentioned different processes has been portrayed in Table 1.

Literature review

As the dyeing processes in the textile industries involve a variety of colorants hence the effluents constitute a wide range of dyestuffs in the form of a mixture. A combination of several dye classes in one mixture could have an effect on the rate of removal and degree of decolorization, making the removal process challenging. The primary reason for this is the presence of diversified functional groups in different classes of dyes. To assess the ability of any removal technique, the synthetic effluents prepared in the laboratory should have the same dye constituents as that of the real effluent sample. These effluents in addition to the dyestuffs also contain other auxiliaries, surfactants and solvents in different amounts. Therefore all the parameters have to be taken into consideration in the removal process.

Physico-Chemical Methods

Various Physico-Chemical techniques are known which can effectively decolorize textile wastewater, but amongst them Adsorption (Crini *et al.*, 2006) 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.

Adsorption

The applied use of Cetyltrimethylammonium bromide (CTAB)-modified TiO_2 nanoparticle as a novel adsorbent for the removal of a mixture of anionic dyes in single and binary systems (Zolgharnein *et al.*, 2014). Isotherm and kinetics of the treatment were also

Table 1: Treatment methods for Textile waste water

S. No.	Techniques
1.	Physico-Chemical Process
	Membrane processes, Reverse Osmosis, Coagulation/flocculation
	Ion exchange, Activated carbon adsorption, Biosorption
2.	Biological Methods
	Bacterial Biodegradation (aerobic and anaerobic),
	Fungal Degradation ,Enzymatic Degradation
3.	Advanced oxidation Process
	Ozonation, Ozone/Hydrogen Peroxide, UV/ozone, Photo catalysis
	Fenton Process, Photo-Fenton, Electrochemical Oxidation
	UV/ozone/hydrogen peroxide, UV/hydrogen peroxide

investigated, in addition to this the use of, nano- -alumina an efficient adsorbent was introduced for removing a mixture of two textile dyes i.e. alizarin red and alizarin yellow and rubber tyre for adsorption for acid blue 113 (Gupta *et al.*, 2011), 2, 4 D removal by using industrial waste (Gupta *et al.*, 2006) and other metal and dyes impurities (Gupta *et al.*, 1986, 1991, 1993, 2000, 2003a, 2003b, 2003c, 2003d, 2005a, 2005b, 2005c, 2006a, 2006b, 2006c, 2006d, 2013, 2014). To achieve higher adsorption capacity and removal percentage, Taguchi design and principle component analysis were applied for the binary mixture of dyes. Several factors were studied through Taguchi technique which indicated the pH to be an influential factor amongst all (Zolgharnein *et al.*, 2014). Researcher evaluated the potential of different adsorbent like Poly vinyl alcohol (PVA)-enhanced hybrid hydrogels of hyperbranched Poly (ether amine) (hPEA) for selective adsorption of mixture of dyes (Deng *et al.*, 2013), chitosan beads also used for decolorization of mixture of three commercial dyes during adsorption treatment. The data was analysed and determined by using spectrophotometer and a multivariate calibration method (Mincea *et al.*, 2013). In continuation to this one research group investigated adsorption of three basic textile dyes in single and multi dye solutions onto bentonite (Turabik and Gozmen, 2013). Similarly removal of binary mixture of dyes by adsorption using tamarind kernel powder over commercial activated carbon (Shanthi and Mahalakshmi, 2012) and a binary mixture of dyes was adsorbed using rarasaponin bentonite, a clay based adsorbent for evaluation of equilibrium data various adsorption isotherms were applied (Kurniawan *et al.*, 2012; Bouatay *et al.*, 2014). Two granular inorganic–organic pillared clays as adsorbents were experimented for basic dyes (Gupta *et al.*, 2000, 2014). These adsorbents were prepared by high shear wet granulation from an Al cetyltrimethylammonium bromide intercalated clay powder (particle diameter < 50 µm) (Cheknane *et al.*, 2010). Parrally decolorization of the different mixture of dyes using bamboo derived activated carbon, In the process Bamboo waste was activated by phosphoric acid at 400 to 600 °C, prediction of experimental data for the binary component system was done using extended single-component equilibrium isothermal models (Chan *et al.*, 2009).

Adsorption kinetics and isotherms for anionic dyes were studied using Polydiallyldimethyl ammonium bentonite for single, binary and ternary system of the dye solutions. For mixed dye solutions, extended Langmuir isotherm was followed and the process proceed through pseudo second order kinetics (Shen *et al.*, 2009). Parallely Poly epichlorohydrin dimethyl amine (EPIDMA) bentonite was used as an adsorbent for the decolorization of single and mixed dye solutions. Different parameters like amount of adsorbent, color removal efficiency, residual color distribution and adsorption kinetics were investigated. Preferential adsorption was observed for the dye having higher affinity for the adsorbent. EPIDMA bentonite exhibited its potential to decolorize anionic dyes both in mixture and individual dye solution. Adsorption was well described by Langmuir isotherm model and followed pseudo second order kinetic model (Kang *et al.*, 2009). Granular activated carbon and silkworm pupa, were tested as adsorbents, silkworm pupa being a natural adsorbent for cationic textile dyes for single and binary solute systems. Equilibrium isotherms were studied for single solute systems and three binary combinations of the dyes. The results favoured the adsorption of synthetic dye solution over granular activated carbon than silk worm pupa. The adsorption on these adsorbents occurred by chemisorption, which remained unaffected by the presence of molecular oxygen (Noroozi *et al.*, 2008). Batch studies were carried out for a binary mixture of dyes onto bentonite using first order derivative spectrophotometric method. Langmuir and Freundlich isotherm models were applied to experimental data and the isotherm constants were calculated for both the dyes. With the increase in the concentrations, the equilibrium uptake amounts of both the dyes in binary mixture onto bentonite decreased resulting in their antagonistic effect. The extended Freundlich isotherm model successfully predicted the multi-component adsorption equilibrium data at moderate ranges of concentration. The process was endothermic and spontaneous in nature (Turabik, 2008).

An agricultural by-product, Rice hull, was tested for its adsorption capability for the adsorption of both basic and reactive dyes. The adsorbent was modified by treating it with ethylenediamine. Sorption was found to be dependent on pH and concentration and equilibrium data could be fitted into both the Langmuir and Freundlich isotherms (Ong *et al.*, 2007). In continuation some scientists study the decolorization

of a mixture of methyl blue, erichrome black T and phenol red dyes using activated charcoal and different parameters were evaluated, adsorption decreased with the increase in pH and temperature. Low temperature was favourable for adsorption process (Iqbal and Ashiq, 2007). Besides these several other wastes like sawdust (Chakraborty *et al.*, 2006), peat (Allen *et al.*, 1988), lignin (Suhas *et al.*, 2007), rubber tire (Al-Saadi *et al.*, 2013; Saleh *et al.,* 2013; Suwanchawalit *et al.*, 2011; Gupta *et al.,* 2011a, 2011b, 2012a, 2012b, 2013) and industrial waste, have also been experimented for a similar reason as adsorbents specifically for synthetic solutions of the dyes. A summary of the some researchers employing adsorption technique for the treatment of mixture of dyes has been portrayed in Table 2.

Biosorption

For degradation of a mixture of dyes, a biosorbent *Portulaca grandiflora* was planted in a tunnel, developed using drilled PVC pipe through which the dye mixture and effluent was made to pass, 87% to 90% decolorization was obtained after 96 hours and 60 hours during treatment of effluent and dye mixture respectively (Khandare *et al.*, 2011). Adsorbtion power of *Saccharomyces cerevisiae* was studied for its biosorption capacity for a mixture of dyes, in which maximum sorption was achieved at optimum pH 6. Various isotherm models were applied and Freundlich isotherm model was found to be the best fit for the biosorption equilibrium data (Ghaedi *et al.*, 2013). A group of researchers explored the phytoremediation potential of *Petunia grandifloraJuss.*, for the degradation of a mixture of dyes and real dye effluent (Watharkar *et al.*, 2013). *Glandularia pulchella* proved to be an good adsorbent, The textile effluent was obtained from a local dyeing industry and a synthetic mixture of dyes was made by adding 10mg of each of the five structurally different dyes to distilled water in order to obtain a final concentration of 50 mg/L. Decolorization extent for the individual dyes was calculated on the basis of initial and final absorbance. For the textile effluent and dye mixture American Dye Manufacturer Institute (ADMI) analysis was used, since the effluent and dye mixture did not show well defined peaks in UV-Visible spectrophotometer, due to the complex structure of dye molecules. *A* reduction in ADMI from 405 to 21 and 418 to 22, in case of textile effluent and dye mixture was observed respectively. High-performance liquid chromatography (HPLC), Fourier transform infrared spectroscopy (FTIR) and

Table 2: Adsorption technique for decolorization of mixture of dyes

Dyes	Decolorization (%)	Reference
Alizarin Red S and Indigo Carmine Dyes	efficient	Zolgharnein *et al.,* (2014a)
Alizarin Red and Alizarin Yellow	99 and 78.5	Zolgharnein *et al.,* (2014b)
Ponceau S/Bismarck brown Y and Orange G/Methylene Blue	-	Deng *et al.,* (2013)
Tartrazine, Amido Black, Congo Red.	-	Mincea *et al.,* (2013)
Basic dyes	-	Turabýk and Gozmen, (2013)
Malachite Green and Methylene Blue Dyes	High	Shanthi and Mahalakshmi, (2012)
Malachite Green and Methylene Blue	High	Kurniawan *et al.,* (2012)
CI Basic Yellow 28 and CI Basic Green 4	-	Cheknane *et al.*, (2010)
Acid Yellow 117 (AY117) and Acid Blue 25 (AB25).	-	Chan *et al.,* (2009)
Acid Scarlet GR(AS-GR), Acid Turquoise Blue 2G (ATB-2G) and Indigo Carmine (IC)	61-71	Shen *et al.,* (2009)
Direct Fast Scarlet, Eosin Y, Reactive Violet K-3R	95	Kang *et al.,* (2009)
Basic Blue 41 and Basic Red 18	-	Noroozi *et al.,* (2008)
Basic Yellow 28 and Basic Red 46	-	Turabik, (2008)
Basic Blue 3 (BB3) and Reactive Orange 16 (RO16)	efficient	Ong *et al.,* (2007)
Methyl Blue, Eriochrome Black T , Phenol Red	100	Iqbal and Ashiq, (2007)
Cibacron Red RB and Cibacron Black B	-	Chakraborty *et al.,* (2006)
Basic dyes	-	Allen *et al.,* (1988)

High Performance Thin Layer Chromatography (HPTLC) analysis confirmed the degradation of dyes from textile effluent and dye mixture (Kabra *et al.*, 2012).

Dead fungal biomass of *Aspergillus niger* has also been used as an efficient biosorbent for the mixture of dyes in aqueous solution. Maximum sorption in this case the optimum pH was found to be 5 and within 10 min maximum decolorization observed for a dye concentration of 20 ppm. Factorial design was applied to determine the effect of different factors on the sorption process. Biosorption increased with increase in pH, dye concentration and biosorbent. Possibility of recycling of biomass was also investigated through desorption by 0.1M HCl and with 50% ethanol in which 0.1M HCl gave better results (Bhole *et al.*, 2004). The two low-cost and renewable biosorbents i.e. apple pomace and wheat straw for the removal of textile wastewater in which the removal was studied using five commercially used dyes. Parameter like effect of dye concentration, size of biosorbent, amount of biosorbent, adsorbance and dye removal were examined for the extent of adsorption (Robinson *et al.*, 2002). In the same year corncob and barley husk were also investigated. Effects of various parameters such as initial dye concentration, particle size, dose of biosorbent, effective adsorbance, and dye removal kinetics was examined for a mixture of five textile dyes (Robinson, 2002) and use three agricultural residues, wheat straw, wood chips and corn-cob shreds for the removal of dyes from a synthetic textile dye effluent. About 75% color was removed from the solution of mixture of dyes (Nigam *et al.*, 2000). It gives a brief information about the dyes decolorized by Biosorption technique Table 3.

Other Physico-Chemical methods

Various other Physico-Chemical methods like flocculation, coagulation, electro-coagulation and membrane filtration techniques have also been employed for decolorization of a matrix of dyes, which have been listed in Table 4. In a different study, Electro coagulation technique was applied for the removal of color from solution containing a mixture of disperse dye and a basic dye. The optimum pH for the treatment was found to be 7 and various parameters like concentration of NaCl and current density were investigated (Sengil and Ozdemir, 2012). The two coagulants i.e. magnesium chloride and ferrous sulphate during coagulation were compared for the treatment of synthetic dye mixture in seven equi-ratio combinations. Results showed $MgCl_2$/Lime to be superior coagulant system on comparison (Verma *et al.*, 2012). Peroxide coagulation process for the removal of mixture of dyes was employed giving 95% of decoloration, the experimentation work was done using an open, undivided and cylindrical glass cell in the presence of sulphate electrolyte media with cathode as a carbon nanotube–polytetrafluoroethylene (CNT–PTFE) (Gupta *et al.*, 2013; Zarei *et al.*, 2010). The efficiency of Electro coagulation in removing color from synthetic and real textile wastewater was tested in which satisfactory results were obtained for both individual and mixed dyes (Phalakornkule *et al.*, 2010).

Residual color profiles of simulated reactive dye wastewater in a chemical flocculation process were determined by a multiple linear regression method, Color removal efficiency was investigated for single dye and the mixture of dyes by using polydiallyldimethylammoniumchloride (PDADMAC), at the optimal dosage of PDADMAC, the color removal

Table 3: Biosorption technique for decolorization of mixture of dyes

Dyes	Decolorization (%)	Reference
Brilliant green (BG) and Methylene blue (MB)	-	Ghaedi *et al.*, (2013)
Remazol Red, Navy Blue HE2R, BBG, Navy Blue RX, Golden Yellow HER, Green HE4BD, Red HE3B, Remazol Orange 3R, Direct Red 5B, and Red M5	-	Watharkar *et al.*, (2013)
Scarlet RR (SRR), Rubine GFL (RGFL), Brilliant Blue R(BBR), Navy Blue 2R(NB2R), Red HE3B (RHE3B)	94.73	Kabra *et al.*, (2012)
Methyl Violet & Basic Fuchsin	100	Bhole *et al.*, (2004)
Five textile dyes	81 - 91	Robinson *et al.*, (2002a)
Five textile dyes	92	Robinson *et al.*, (2002b)
Synthetic mixture of dyes	70-75	Nigam *et al.*, (2000)

Table 4: Other physico- chemical techniques for decolorization of mixture of dyes

Dyes	Decolorization (%)	Reference
Mixture of Blue disperse and Yellow basic	-	Sengil and Ozdemir, (2012)
Two diazo dyes namely Reactive Black 5 (RB5) and Congo Red (CR) and one anthraquinone dye as Disperse Blue 3 (DB3)	81-99	Verma *et al.*, (2012)
C.I. Basic blue 3 (BB3), Malachite green (MG), C.I. Basic red 46 (BR46) and C.I. Basic Yellow 2 (BY2)	90	Zarei *et al.*, (2010)
Blue reactive dye (Reactive Blue 140), Disperse dye (Disperse Red 1).	>95	Phalakornkule *et al.*, (2010)
Reactive Red K-2BP (K-2BP), Reactive Violet K-3R (K-3R) and Reactive Black KN-B (KN-B)	>90	Kang, (2007)
KN-B and Reactive Violet K-3R (K-3R), Reactive Red K-2BP (K-2BP)	90	Kang *et al.*, (2007)
Reactive black dye (Cibacron Black B) and reactive red dye (Cibacron Red RB).	92-94	Chakraborty *et al.*, (2003)

efficiencies for the mixture of dyes were greater than 90%. A multiple linear regression method was used to determine residual color profiles of mixture of reactive dyes during a chemical flocculation process. A single, binary and ternary dye mixture solution was decolorized using poly epicholorohydrin-diamine (PEPIDA) (Kang, 2007). Textile effluents have also been decolorized by the use of membrane based separation process like Nanofiltration. The experiment on dye mixture was carried out in an unstirred batch and a rectangular cross flow cell and 94% of Chemical Oxygen Demand (COD) removal was observed (Chakraborty *et al.*, 2003).

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 utilised to remove color of a wide range of dyes through anaerobic, aerobic, and sequential anaerobic-aerobic treatment processes (Gupta *et al.*, 2010a, 2010b).

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 non-

toxic degraded products (Kolekar *et al.*, 2013). Some other strains of bacteria like *Pseudomonas fluorescens* strains (Sz6 and SDz3) (Godlewska *et al.*, 2014) and *Shewanella* bacterial strains (Liu *et al.*, 2013) 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 (Saratale *et al.*, 2013). 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 (Kabra *et al.*, 2013). An immobilized bacterial consortium was also used for the aerobic biodegradation of mixture of sulphonated azo dyes like acid orange 7 and acid red 88. The treatment was done in a two stage i.e. sparged packed and bed biofilm reactor for better results (Vasconcelos *et al.*, 2012). *Aeromonas hydrophila* a bacterial strain was isolated and identified for degrading a mixture of dyes. Anoxic culture was found to be the best for color removal at 5.5-10.0 pH and optimum temperature of 20-30ºC (Naik and Singh, 2012). Decoloration and physico-chemical parameters of dye mixture were analysed by using *Bacillus*, a type of bacteria which proved to be efficient in color and

COD removal (Jayan *et al.*, 2011). A mixture of seven commercial dyes having different structures were degraded by an eco-friendly strain *B. laterosporous* in which 87% ADMI color removal was obtained in 24 hours at an optimum temperature of 40 C, with 71% COD and 49% total organic carbon (TOC) reduction. Experiments were carried out by varying different parameters like pH, temperature and presence of different salts. Phytotoxicity studies revealed that less toxic metabolites were produced after the degradation of mixture of dyes (Kurade *et al.*, 2011).

The ability of *Sphingo bacterium* sp. ATM was tested to degrade a mixture of dyes and textile efûuent. Three combinations of mixtures containing five dyes (mixture 1), seven dyes (mixture 2), and ten dyes (mixture 3) were prepared having 0.5 g/L concentration. The decolorization of mixture of dyes and textile efûuent was calculated using ADMI tristimulus ûlter method. Among all the tested microorganisms, isolated *Sphingo bacterium* sp. ATM effectively decolorized the dye direct blue (100%) GLL and simultaneously produced polyhydroxyhexadecanoic acid (64%). The organism produced approximately 66% and 61% polyhydroxyhexadecanoic acid while decolorizing the dye matrix and textile efûuent respectively. There was a signiûcant reduction in the COD and BOD (Biological Oxygen Demand). The optimum temperature for dye decolorization was found to be 37°C and the rate of decolorization decreased with the increase in temperature (Tamboli *et al.,* 2010). A mixture of eight textile dyes was also degraded using bacteria where the ADMI value for dye removal was 89% and 67% for dye mixture and industrial effluent, respectively at optimum pH and temperature of 5 and 50°C, respectively (Joshi *et al.*, 2010). The consortium SKB-II consisted of five different bacterial types identified by 16S rDNA sequence alignment as *Bacillus vallismortis*, *Bacillus pumilus*, *Bacillus cereus*, *Bacillus subtilis* and *Bacillus megaterium* were used for the decolorization of individual dyes and the synthetic mixture of dyes (Tony *et al.*, 2009). In 2008 experiment was carried out using three bacterial strains *Aeromonascaviae*, *Proteus mirabilis* and *Rhodococcus globerulus* for azo dyes (Joshi *et al.*, 2008), in addition to this AS96 (*Shewanella putrefaciens*) strains were also found to be effective in the decolorization process (Khalid *et al.*, 2008). Table 5 clearly depicts the bacterial biodegradation technique used for decolorizing dye mixture.

Newly isolated halophilic and halotolerant bacteria (Asad *et al.,* 2007) from coconut coir sample (Vijaykumar *et al.*, 2007) worked efficiently for removal of dyes from mixture. Some newly isolated strains from sludge samples and mud lakes also could efficiently decolorize the mixture of dyes. The optimum pH and temperatures were pH 5.5-10.0 and 20°C-35.8°C. This strain effectively decolorized the media containing a mixture of dyes (24 dyes) within two days of incubation (Chen *et al.*, 2003). Azo dyes have also been further decolorized by the use of some immobilised bacteria by using PVA (phosphorylated polyvinyl alcohol) (Chen *et al.*, 2003). The efficiency of consortia for the removal of azo dye and simulated wastewater degradation in presence of various co substrates, Potential microorganisms identified were *Pseudomonas sps*, *Bacillus sphalomonas sps*, *Orthrobacter sps*, *Micrococci sps*, which degraded the mixture of commercial azo dyes (Padamavathy, 2003). In 1999 *Kurthia sp.* was screened on the basis of rapid dye decolorizing activity for triphenylmethane dyes. Aerobically this strain was able to decolorize 98% of color intracellularly. A number of triphenylmethane dyes were studied individually and in mixture. The rates of decolorization were different for different dyes ranging from 92-100%, except for ethyl violet (8%) which was used. For synthetic mixture of dyes 98% decolorization was obtained which was more than that obtained for textile dye-stuff effluent (56%) (Sani *et al.*, 1999). A combination of azo, diazo and reactive dyes was also studied (Nigam *et al.*, 1996).

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 (Taha *et al.*, 2014). 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 (Przystas *et al.*, 2013). Performance of immobilized *Phanerochaete chrysosporium* in modified PVA matrix was investigated for decolorization of mixture of textile dyes, Central composite design was used to determine optimum operating conditions (Idris *et al.*, 2013).

Table 5: Bacterial biodegradation of mixture of dyes

Dyes	Decolorization (%)	Reference
Raspberry red, Golden yellow HER, Orange 2R, Orange M2R	-	Kolekar *et al.,* (2014)
Diazo Evans blue (EB) and triphenylmethane brilliant green (BG).	88.9	Zab³ocka-Godlewska *et al.,* (2014)
Mixture of dyes	87 (ADMI)	Saratale *et al.,* (2013)
Amaranth, Acid Orange 52, Direct Blue 71	efficient	Liu *et al.,* (2013)
Brown 3REL (B3REL), SRR, Remazol Red (RR), Direct Red 2B (DR2B)and MalachiteGreen(MG)	100	Kabra *et al.,* (2013)
Sulphonated Azo dyes, Acid Orange 7 (AO7) and Acid Red 88 (AR88)	~100	De los Cobos Vasconcelos *et al.,* (2012)
Mixture of dyes	high	Naik *et al.,* (2012)
Orange P3R, Yellow P3R, Blue H5R, Violet P3R, Brown P5R, Black V3R, Orange P2R	47.24	Jayan et al., (2011)
Remazol Red, Rubine GFL, Brown 3REL, Scarlet RR, Golden Yellow HER, Methyl Red, Brilliant Blue GL	87	Kurade *et al.,* (2011)
Mixture 1: Navy Blue Rx, Navy Blue 3G, Blue 2RNL, Golden Yellow HER, and Green HE4B.Mixture 2 :Red HE8B, Red M5B, Remazol Red, Orange HE2R, Golden Yellow HER, Green HE4B, and Direct Blue GLL,Mixture 3:Orange HE2R, Golden Yellow HER, Green HE4B, Navy Blue Rx, Navy Blue 3G, Blue 2RNL, Direct Blue GLL, Red HE8B, Red M5B, Remazol Red.	Mixture 1 64.89 Mixture 2 72.88 Mixture 3 61.4	Tamboli *et al.,* (2010)
8 textile dyes	89	Joshi *et al.,* (2010)
Congo red, Bordeaux, Ranocid Fast Blue and Blue BCC	50-60	Tony *et al.,* (2009)
16 Azo dyes	90	Joshi *et al.,* (2008)
Acid Red 88, Reactive Black 5, Direct Red 81, and Disperse Orange 3	100	Khalid *et al.,* (2008)
Azo dyes	-	Asad *et al.,* (2007)
Naphthalene-containing sulfonated azo dyes Amaranth , Fast Red E and Ponceau S	100	Vijaykumar *et al.,* (2007)
Acid Blue 74, Acid Orange 7, Acid Red 106, Direct Yellow 4 and Direct Yellow 12 (Acid Black 172, Acid Blue 264, Acid Yellow 42, Direct Black 22, Direct Orange 39, Direct Red 224, Direct Red 243, Direct Yellow 86, Reactive Black NR, Reactive Black 5, Reactive Blue 160, Reactive Blue 171, Reactive Blue 198, Reactive Blue 222, Reactive Green 19, Reactive Red 120, Reactive Red 141, Reactive Red 198 and Reactive Yellow 84)	90	Chen *et al.,* (2003a)
Everzol Red RBN (Reactive Red 198a; R-r198b) , Everzol Yellow 3RS (Reactive Yellow 176a; R-y176b) , Everzol Black B (Reactive Black 5a; R-BK5b) , Everzol Brilliant Red 3BS (Reactive Red 239a; R-r239b) ,Everdirect Supra Red BWS (Direct Red 243a; D-r243b) ,Everdirect Supra Yellow RL (Direct Yellow 86a; D-y86b) , Everdirect Supra Orange 2GL (Direct Orange 39a; D-o39b) , Sigma Orange (acid orange 7a; A-o7b	>95	Chen *et al.,* (2003b)
Red RB, Remazol Red, Remazol Blue, Remazol Violet, Remazol Yellow, Golden Yellow, Remazol Orange, Remazol Black	95	Padmavathy *et al.,* (2003)
Magenta, crystal violet, pararosaniline, brilliant green, malachite green, ethyl violet	98	Sani *et al.,* (1999)
Cibacron Red (reactive dye), Remazol Golden Yellow (azo dye) , Remazol Red (diazo dye); Remazol Navy Blue (diazo dye) and Cibacron Orange (reactive dye) and Remazol Blue	100	Nigam *et al.,* (1996)

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 (Ambrosio *et al*., 2012), 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 (Waghmode *et al*., 2011).

Synthetic wastewater containing two azo dyes was decolorized both taking individual and a combination of dyes using immobilized *Phanerochaete chrysosporium* in a batch-operated rotating biological contactor (RBC) reactor (Pakshirajan *et al.*, 2010). The use of white rot fungus *Bjerkandera sp.* BOL 13 for the decolorization of dyes in batch and continuous modes, a continuous experiment was performed to decolorize mixture of three dyes (Nordstrom *et al.*, 2008). The potential of *Trametes villosa* and *Pycnoporus sanguineus* was explored in the decolorisation of a mixture of ten reactive textile dyes, dye decolorization was faster in the mixed culture (two fungi) compared to the pure culture, with approximately 80% and 90% color reduction on seventh and seventeenth day, respectively (Machado *et al*., 2006). Similarly, decolorization of the mixtures of different reactive textile dyes by the white-rot basidiomycete *Phanerochaete sordida* and studied the inhibitory effect of poly vinyl alcohol, white-rot basidiomycete *Phanerochaete sordida* decolorized dye mixtures (200 mg/L each) by 90% within 48 h in nitrogen-limited glucose–ammonium media. Decolorization of dye mixtures needed Mn^{2+} and Tween 80 in the media indicating a major role play of Manganese (Harazono and Nakamura, 2005). Azo dyes with different structures were investigated for decolorisation by the use of *Cunning hamellaelegans* (fungi) under several media conditions. It was observed that the factors on which decolorization depended were incubation time, molecular structure of the dye and the presence or absence of co-substrates. Spectrophotometeric

studies revealed the occurrence of biodegradation along with biosorption (Ambrosio and Takaki, 2004).

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 (Amaral *et al*., 2004). A group of researchers evaluated the possibility of a fungal wastewater treatment for a mixture of bioaccessible reactive azo dyes using biodegradation assays (Martins *et al*., 2003). White-rot fungus *Ph. tremellosa* was found capable of decolorizing an array of synthetic textile dyes (Kirby *et al.*, 2000). A 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. The process was found to be time dependent and followed first order kinetics corresponding to the initial concentration of dyes. Investigations revealed that the process required a biodegradable substrate such as glucose (Sumathi and Manju, 2000). Table 6 depicts some of the different dye mixtures decolorized by fungal degradation.

Enzymatic methods

Ammonium sulphate fractionated pointed gourd (*Tricho santhesdioica*) peroxidase- concanavalin A (PGP-Con A) complex, entrapped into calcium alginate-pectin 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 (Jamal *et al*., 2013). 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 (Zeng *et al.*, 2012). A similar degradation of a mixture of reactive dyes was carried out by using commercial laccase in a batch reactor, mathematical models were

Table 6: Fungal degradation of mixture of dyes

Dyes	Decolorization (%)	Reference
Azo–Anthraquinone dye mixture (Azure B, Congo Red, Trypan Blue and Remazol Brilliant Blue R)	74.93	Taha *et al.,* (2014)
Triarylmethane dye (Brilliant green) and Diazo dye (Evans blue)	80	Przystas *et al.,* (2013)
Yellow FG, Red 3BS, Orange 3R, Blue RSP, Black B, and remazol turquoise blue	82	Idris *et al.,* (2013)
Reactive azo dyes (red, black and orange II)	88	Ambrósio *et al.,* (2012)
Remazol Red, Golden Yellow HER, Rubine GFR, Scarlet RR, Methyl Red, Brown 3REL, Brilliant Blue	88	Waghmode *et al.,* (2011)
Direct Red-80 and Mordant Blue-9	77–97	Pakshirajan and Singh (2010)
Reactive blue 21, Reactive black 5 and Reactive orange 13	60-66	Nordstrom *et al.,* (2008)
Remazol Brilliant Orange 1, Levafix Gold Yellow 10, Procion Yellow 14, Drimaren Brilliant Blue 17, Remazol Brilliant Blue 18, Cibacron Black 55, Procion Black 59, Drimaren Turquoise Blue 62, Drimaren Brilliant Red 67, and Remazol Red 75.	80-90	Machado *et al.,* (2006)
Mixtures of 4 reactive textile dyes, azo and anthraquinone dyes	90	Harazono and Nakamura, (2005)
Orange , Reactive Black , Reactive Red	88	Ambrosio and Campos-Takaki, (2004)
ProcionOrange MX-2R (C.I. Reactive Orange 4), Remazol Red 3B (C.I. Reactive Red 23) and Remazol Black GF (C.I. Reactive Black 5)	97	Amaral *et al.,* (2004)
Meta or para aminobenzoic or aminosul-Phonic acids as diazo components and two fungal bioaccessible groups present in lignin structure, guaiacol or syringol, as coupling components	100	Martins *et al.,* (2003)
Cibacron Red, Remazol Navy Blue ,Remazol Red , Cibacron Orange , Remazol Golden Yellow , Remazol Blue ,Remazol Turquoise Blue ,Remazol Black B	>96	Kirby *et al.,* (2000)
Drimarene dyes.	95	Sumathi and Manju, (2000)

developed to simulate the kinetics of laccase catalysed degradation by reactive dyes in mixture (Cristovao *et al.*, 2009). 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 (Chhabra *et al.*, 2008). 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. The optimum pH for decolorization of the complex mixture was observed to be 3.0. On increasing the concentration of enzymes decoloration was also observed to increase (Khan and Husain, 2007). Enzymatic method used for decolorizing different mixtures of dyes has been shown in Table 7.

Salt fractionated turnip *(Brassica rapa)* peroxidases was used in another research to degrade acid dyes. The selected enzymes were able to decolorize most of the acid dyes in the presence of 2.0 mM 1-hydroxybenzotriazole (HOBT). Dye decoloration was found not to be affected by an increase in the concentration of enzyme and time, in the absence of HOBT. Optimum pH was 5.0 and temperature 40 °C. Complex mixtures of dyes were decolorized with an enzyme in the presence of HOBT (Kulshrestha and Husain, 2007). Experiments

Table 7: Enzymatic degradation of mixture of dyes

Dyes	Decolorization (%)	Reference
Disperse Red 19 & Disperse Black 9	82.1	Jamal *et al.,* (2013)
Azo and Anthraquinone dyes	-	Zeng *et al.,* (2012)
Reactive Black 5, Reactive Yellow 15, Reactive Red 239	Appreciable Decolorization	Cristovao *et al.,* (2009)
Simulated dye mixture (reactive & acidic dyes :Reactive Orange Reactive Red 198)	80	Chhabra *et al.,* (2008)
Reactive Blue 4, Reactive Blue 160, Reactive Blue 171, Reactive Red 11, Reactive Red 120, Reactive Orange 4, Reactive Orange 86 and Reactive Yellow 84	87 – 98	Khan and Husain, (2007)
Mixture of dyes	Significant	Kulshrestha and Husain, (2007)
Direct dyes	80	Matto and Husain, (2007)
Mixture of three, four and eight textile dyes	80	Akhtar *et al.,* (2005)

demonstrate the effect of salt fractionated turnip proteins for direct dyes using different redox mediators, maximum decoloration was obtained when a complex mixture of dyes was tested with the enzyme in presence of 0.06 mM redox mediator (1-hydroxybenzotriazole/violuric acid) (Matto and Husain, 2007). In a different study on textile waste water immobilized bitter gourd *(Momordica charantia)* peroxidase was used for the decolourization and was found highly effective. The optimum pH was in the range of pH 3.0–4.0 at 40°C. Mixtures of three, four and eight dyes was prepared and treated with soluble and immobilized bitter gourd peroxidase. Each mixture was decolorized by more than 80% when treated with immobilized enzyme (Akhtar *et al*., 2005).

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 (Gupta *et al*., 2012). 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 (Kestioglu *et al*., 2005).

Various advanced oxidation methods have been employed for removal of mixture of dyes.

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 (Wijannarong *et al.,* 2013). Synthetic dye effluent was prepared using nine commercially reactive dyes and similar ozonation was carried out (Sancar and Balci, 2013). Table 8 reports the ozonation technique used for decolorization of matrix of dyes. 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. Simple by products like chlorides, sulphates, nitrates were formed as a result

Table 8: Ozonation for decolorization of mixture of dyes

Dyes	Decolorization (%)	Reference
Nova cron super black G and Terasil red ww 3BS	90	Wijannarong *et al.*, (2013)
Nine commercial reactive dyes Monoazo, diazo, azo-cu dyes	effective	Sancar and Balci, (2013)
Remazol Red (RR), Red RB(RRB), Remazol Blue (RB), Remazol Black (Rbla), Remazol Violet (RV), Remazol Orange (RO), Golden Yellow (GY), Remazol Yellow (RY)	95-99	Sarayu *et al.*, (2007)

of oxidation and cleavage of substituent groups present in dye molecules (Sarayu *et al.*, 2007).

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 (Chatzisymeon *et al.*, 2006). A mixture of nine reactive dyes from azo, anthraquinone and triazine classes of dyes were degraded in chlorine mediated electrochemical oxidation. Complete decolorization was obtained with high removal of COD at 120 minutes

of the treatment (Rajkumar and Kim, 2006). Electrochemically decolorized dyes by means of DC electric current. 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 (Sanroman *et al.*, 2004). Table 9 provides a brief account of the researches carried out for dye mixtures using Electrochemical Oxidation method.

Photo Catalytic Treatment

In this process photo degradation of a mixture of two textile dyes using TiO_2 catalysed microsphere. Results suggests microsphere as a promising catalyst for wastewater treatment (Li *et al.*, 2014). Similarly CuO-clinoptilolite was used for the photodecolorization of mixture of methylene blue and bromophenol blue (Ejhieh and Mobarakeh, 2014). Photo degradation of a mixture of the two cationic azo dyes was studied using CuO/nano-zeolite catalyst under solar irradiation, results showed that decolorization kinetics fitted well to the Langmuir–Hinselwood first order rate law

Table 9: Electrochemical oxidation for decolorization of mixture of dyes

Dyes	Decolorization %	Reference
Remazol Black B, Remazol Red RB, Remazol Golden Yellow RNL, Cibacron Red FN-R, Cibacron Red FN-G, Cibacron Black WNN, Drimaren Red K-8B, Drimaren Scarlet K-2G, Drimaren Yellow K-2R, Drimaren Navy K-BNN, Drimaren Yellow K-4G, Drimaren Orange X-3LG, Drimaren Blue X-3LR, Drimaren Violet K-2RL, Drimaren Red K-4BL, Drimaren Blue K-2RL	100	Chatizisymeon *et al.*, (2006)
Reactive Red 120, Reactive red 141, Reactive red 198, Reactive blue 49, Reactive blue 19, Reactive orange 16, Reactive black 5, Reactive yellow 84, Reactive yellow 15	100	Rajkumar and Kim, (2006)
Bromophenol Blue, Indigo, Poly R-478, Phenol Red, Methyl Orange, Fuchsin, Methyl Green and Crystal Violet	100	Sanromán *et al.*, (2004)

(Ejhieh and Shamsabadi, 2013). A photo decolorization of a mixture of textile dyes viz. methyl orange and bromocresol green in presence of a heterogenous catalyst. CuS incorporated clinoptilolite zeolite was used as a catalyst during photodegradation under sunlight (Ejhieh and Moazzeni, 2013). Photooxidation of mixture of three textile dyes by UV/H$_2$O$_2$ process was reported, for optimization of the process central composite design was applied and degraded products were identified by the Gas chromatography–mass spectrometry (GC–MS) technique (Khatee *et al.*, 2012). The Bismuth vanadate and cerium dioxide (BiVO$_4$/CeO$_2$) nano composites were prepared and used as visible-light photocatalysts in the degradation of a mixture of methylene blue and methyl orange dyes (Wetchakun *et al.*, 2012). With the same purpose, TiO$_2$ as photocatalyst was used during photodegradation of a binary mixture of textile dyes.

The experiment was carried out at different temperatures to calculate rate constant (Shanthi and Priya, 2012). The synthesized nano particles of zinc oxide by co-precipitation technique, the nano particles were used during photocatalytic degradation of a mixture of crystal violet and aniline blue dyes from aqueous solutions (Shanthi and Padmavathi, 2012; Nagda and Ghole, 2008).

Table 10 summarizes the researches on photocatalytic degradation technique for decolorization of a mixture of dyes. Photocatalytic decolorization which is also one of the advanced oxidation processes was carried out for a mixture of two anionic azo dyes (Sharma *et al.*, 2012). Similarly, photocatalytic degradation of a mixture of six commercial azo dyes was investigated by exposure to UV radiation in an aqueous solution. Response surface methodology, based on a 3² full factorial experimental

Table 10: Photo catalytic decolorization of mixture of dyes

Dyes	Decolorization (%)	Reference
Rhodamine B and 4-(*P*-Nitrophenylazo)Resorcinol (Magneson I)	High	Li *et al.*, (2014)
Methylene blue and Bromophenol blue	61 and 32	Nezamzadeh-Ejhieh and Zabihi-Mobarakeh, (2014)
Methylene blue and Rhodamine B	-	Nezamzadeh-Ejhieh and Karimi-Shamsabadi, (2013)
Methyl Orange and Bromocresol Green	-	Nezamzadeh -Ejhieh and Moazzeni, (2013)
CI Basic Red 46 (BR46), Malachite Green (MG) and CI Basic Blue 3 (BB3)	-	Khataee *et al.*, (2012)
Methylene Blue and Methyl Orange	90-98	Wetchakun *et al.*, (2012)
Aniline blue and Crystal Violet	-	Shanthi and Priya, (2012)
Crystal Violet and Aniline Blue	100	Shanthi and Padmavathi, (2012)
Two anionic azo dyes: Reactive Red 120 and Reactive Black 5	-	Sharma *et al.*, (2012)
Six Azo dyes	100	Palacio *et al.*, (2012)
Remazol Black (RB5) and Red Procion MX-5B	-	Sahel *et al.*, (2010)
Acid Orange 7 (AO7) and Reactive Red 2 (RR2)	45-60	Juang *et al.*, (2010)
Basic Red 46 and Basic Yellow 28	-	Gözmen *et al.*, (2009)
Binary mixture of reactive dyes	90-95	Chatterjee *et al.*, (2008)
Crystal Violet (CV), also known as C.I. Basic Violet 3, which is a cationic triphenylmethane dye and Methyl Red (MR), which is an anionic azo dye	99	Gupta *et al.*, (2006)
Bright Blue Remazol (blue reagent-19), Red Procion H-E7B	100	Costa *et al.*, (2004)

design was applied taking various parameters into consideration. Under optimal reactor conditions monitoring of dye mineralisation was carried out. 100% decolorization was obtained after 240 mins of irradiation which also followed pseudo-first kinetic order model (Palacio *et al.*, 2012). A similar process was carried out for a mixture of remazol black (RB5) and red procion MX-5B in presence of TiO_2 Degussa P-25 by UV–VIS spectroscopy (Sahel *et al.,* 2010). Photodegradation under UV irradiation in TiO_2 suspensions of a binary mixture of dyes was also examined. Several factors were considered during the experiment (Juang *et al.*, 2010). A similar Photocatalytic degradation and mineralization of textile dyes in single and binary solutions was performed by $UV/TiO_2/periodate$ system (Gözmen *et al.,* 2009).

Visible light-irradiated TiO_2 semiconductor photo catalyst was used for a binary mixture decolorization (Chatterjee *et al.*, 2008). Crystal violet (basic violet 3) and methyl red dye were photo catalytically degraded in aqueous suspensions using Ag^+ doped TiO_2. The degradation of the dyes in the mixture was measured spectrophotometrically and color removal was found to be >99% on UV irradiation for 90 min in the presence of Ag^+ doped TiO_2 and about 70% under similar conditions in presence of TiO_2. COD removal was measured at regular intervals to quantify the mineralization of the dye. Above 86% mineralization was observed using 90 min irradiation. The degradation of dyes was studied spectrophotometrically and results showed that kinetics fitted well to the Langmuir–Hinselwood pseudo first order rate law. Investigation was done to find the effects of pH and different interfering substances like Cl^-, NO_3^-, SO_4^{2-}, HPO_4^{2-}, Ca^{2+}, Fe^{2+}, humic acid, O_2 and electron acceptors like H_2O_2, $(NH_4)_2S_2O_8$ and $KBrO_3$ (Gupta *et al.*, 2006). Also Bleaching and photodegradation of textile dyes by H_2O_2 and solar or ultraviolet radiation was investigated. The photooxidation of a textile dyes mixture of bright blue remazol (blue reagent-19), red procion H-E7B dyes was experimented and the efficiency of photooxidation was compared using hydrogen peroxide (30%) as a bleaching reagent, solar and ultraviolet radiation, common glass borosilicate, quartz assay tubes, and no solid catalysts. The color of a mixture of blue and red dyes was almost completely removed after 3 hours of exposure to solar or ultraviolet radiation (Costa *et al.,* 2004).

Photo-Fentons method

Degradation of mixture of three reactive dyes was studied by electro-Fenton process. In the process carbon paper cathode modified with carbon nanotubes was used. Treatment data was studied by using Partial least square method. Central composite design and ANOVA technique was used during modelling and optimizing the decolorization process (Djafarzadeh *et al.,* 2014). The application of photo-Fenton process $(UV/Fe^{3+}/H_2O_2)$ for treating a mixture of three cationic dyes. Effect of initial concentration of oxidants i.e., Fe^{+3} and H_2O_2 on the dyes degradation was investigated (Chergui *et al.*, 2012). Synthetic textile waste water was degraded with homogenous and heterogenous photo-Fenton oxidation method and results were compared. Synthetic waste water was prepared by mixing remazol red RR and remazol blue RB. Complete decolorization was obtained with homogenous oxidation which showed a higher efficiency than heterogenous photo-fenton method (Punzi *et al.*, 2012). Some researcher studied the treatment of mixture of three azo dyes using homogeneous photoelectro-Fenton (PEF) combined with heterogeneous photocatalytic process (UV/TiO_2). Central composite design was employed for the optimization of photoelectro-Fenton/photocatalytic decolorization of the dye solution containing three dyes using a combined approach. The factors investigated were the initial concentration of three dyes, initial amount of Fe^{3+} and reaction time. Results indicated that the optimum Fe^{3+} concentration was 0.15 mM for decolorization process. Analysis of variance (ANOVA) showed that predicted values matched the experimental values reasonably (Khatee *et al.*, 2012).

A group of researchers employed Electro-Fenton technique for decolourization of dyes in a continuous bubble reactor. Investigation was done for various parameters like iron dosage and pH which play a major role in increasing the efficiency. At optimal conditions several dyes were decolorized using this technique. For dye mixture about 43% of decolorization was obtained after 21 hours with TOC reduction of 46 (Rosales *et al.,* 2009). The analyses of overall kinetics of a model mixture of reactive dyes by Fenton and photo-Fenton oxidation, Investigation was also done for decolorisation, effect of temperature and mineralisation. Batch experiments were performed and the obtained data was used to perform kinetic studies.

Rate constants and half-life time were determined according to a pseudo-first-order degradation kinetic model. The results showed that the Fenton process under solar light was most effective. They also suggested that Fenton and photo-Fenton-type reactions are viable techniques for the treatment of dye mixtures having higher color intensity, chemical oxygen demand and total organic carbon removal (Torrades *et al.*, 2008). A possibility of Fenton-like reagent (Fe^{3+}/H_2O_2) was also tested to decolorize a dye mixture of malachite green and orange II. Investigation on various parameters like Fe^{3+} concentration and initial dye concentration was done which showed that removal efficiency increased by increasing the Fe^{3+} concentration (Rastegar *et al.*, 2008). Another oxidation process known as electro-Fenton was used for a mixture of three frequently used in textile industries. Results showed that the electro-Fenton process quickly degraded the synthetic dye mixture (Lahkimi *et al.*, 2007). The study of the Fenton oxidation process with different combination of dyes considering their actual application doses in dye houses. The optimum pH using $FeSO_4$ and H_2O_2 was 2.5 at 30°C resulting in more than 96% COD and 99% Pt-Co unit of color removal for the mixture of dyes. For mixture of dyes (RB5 + RR + RB + RY) the optimum conditions observed were 4.0 pH at 50°C (Meric *et al.*, 2003). Fenton method used for treating dye matrix has been listed in Table 11.

Combined Techniques

Different methods described in the sections above encourage the use of single techniques for dye discoloration and removal. However if the efficiency factor of all these processes is considered only very few of them show 100% efficiency. Therefore several researches have also been carried out with a combination of different techniques (Gomes *et al.*, 2010) (Table 12).

A combination of removal techniques like Coagulation and Catalytic oxidation was examined for reducing the concentration of mixture of direct dyes. The model solution of mixture of dyes was first coagulated and then oxidized with hydrogen peroxide in presence of Fe(II) or UV as catalyst (Gonta *et al.*, 2014). Some scientists investigated adsorption of a simulated mixture of dyes and real textile effluent using pre-treated sugarcane bagasse (Gupta *et al.*, 1998a, 1998b, 1999, 2000, 2001, 2002, 2003, 2004, 2013) which was further decolorized by solid state fermentation using *Providensia staurti* strain E*bt*SPG. Sugarcane baggase was pretreated with $CaCl_2$, alkali, ammonia, steam and milling amongst which $CaCl_2$ showed 92% and 86% adsorption of simulated dyes mixture and real textile effluent (RTE) respectively (Kadam *et al.*, 2011). In the same year, photo electro adsorption (PEA) decolorization of a mixture of three azo dyes, with polyaniline-modified electrode as a conductive

Table 11: Fenton / photo fenton treatment for decolorization of mixture of dyes

Dyes	Decolorization (%)	Reference
C. I. Reactive Red 195, C. I. Reactive Yellow 84 and C. I. Reactive Blue 69	-	Djafarzadeh *et al.,* (2014)
Cationic dyes: Basic Blue 41, Basic Yellow 28 , Basic red 46	100	Bouafia-Chergui *et al.,* (2012)
Remazol Red RR, Remazol Blue RR	100	Punzi *et al.,* (2012)
C.I. Acid Yellow 36 (AY36), C.I. Acid Red 14 (AR14) and C.I. Basic Yellow 28 (BY28)	-	Khataee *et al.,* (2012)
Methyl Orange, Reactive Black 5 and Fuchsin Acid , Lissamine Green	43	Rosales *et al.,* (2009)
Mixture of reactive dyes	High	Torrades *et al.,* (2008)
Malachite Green and Orange II	-	Rastegar *et al.,* (2008)
Yellow drimaren, congo red and methylene blue	90	Lahkimi *et al.,* (2007)
Remazol Black 5 (RB5), Remazol Red RB (RR), Remazol Yellow 84 (RY), Remazol Brilliant Blue (RB)	99	Meriç *et al.,* (2003)

Table 12: Decolorization of mixture of dyes by combined techniques

Technique	Dyes used	Decolorization (%)	Reference
Coagulation and Catalytic Oxidation With Hydrogen Peroxide	Direct dyes (Direct blue k , Direct brown)	-	Gonta *et al.,* (2014)
Adsorption Followed By Biodegradation	Simulated dye mixture , Real textile effluent	92 , 86	Kadam *et al.,* (2013)
Photo Electro Adsorption	Acid Red 88 (AR88), Acid Blue 92 (AB92), and Acid Orange 2 (AO2)	96	Pirkarami *et al.,* (2013)
Ozone And Electron Beam Irradiation	Mixture of dyes	high	Emami-Meibodi *et al.,* (2013)
Phytoremedation With Bacterial Augmentation	Disperse Red BF, Disperse Yellow G, Disperse Bryal Blue, Rubine GFL, Brown REL	89	Khandare *et al.,* (2013)
Adsorption Followed By Biological Method	Disperse Blue 2BLM, Reactive Yellow XL, Reactive Red RB, Reactive Red ME4BL, Reactive Orange 3R, Reactive Blue ME2RL, Reactive Golden Yellow HER	Adsorption - 62 By Consoritum - 82	Kadam *et al.,* (2011)
Electrocoagulation / Electroflotation	Disperse dye Yellow terasil 4G, Red terasil 343 150% and Blue terasil 3R02, the reactive dye is a mixture of Red S3B 195, Yellow SPD, Blue BRFS	70-90	Balla *et al.,* (2010)
Electrochemical Photodegradation	Methyl Orange, methylene Blue, Reactive Blue 2, Naphthol Blue Black, Direct Rd 81, Rhodamine 6G	>50	Zainal *et al.,* (2007)
Adsorption And Biodegradation	Telon Red M-3B (C.I. Acid Red 131; Azo Dye), Telon Blue M-RLW (C.I. Acid Blue 204; Anthraquinone Dye) and Telon Yellow M-4GL (C.I. Acid Yellow 79	89±1.86	Somasiri *et al.,* (2006)
Biological-Chemical Method	Tartrazine, Sunset Yellow, Reactive Black 5, Drimarene Red, Remazol Brown, Drimarene Navy dyes	80- 95 in 1-24hrs	Patel and Suresh, (2006)
Coagulation And Adsorption	C.I. Reactive Red 45 and C.I. Reactive Green 8	100	Papic *et al.,* (2004)

polymer was reported. The electro polymerised electrode was used in a non-continuous reactor using UV irradiation for the decolorization of mixture of azo dyes at operational conditions. Optimum pH for decolorization was found to be 5 at which maximum color was removed in 40 minutes (Pirkarami *et al.,* 2013). Combined effects of ozone and electron beam radiation for removal of color of dye mixture was studied in which results indicated that higher decolorization was obtained with ozonation after irradiation (Meibodi *et al.,* 2013).

One group employed phytoremediation followed by bacterial degradation. *Portulaca grandiflora* (Moss-Rose) was used in a phyto reactor whose efficacy was increased by augumenting with *Pseudomonas putida* culture. The bacterial consortium and plant was found efficient for the removal of dye mixture. The experimental data was also analysed by ANOVA and Tukey-Kramer comparision Test (Khandare *et al.,* 2013). Decolorization of adsorbed textile dyes by developed consortium of *Pseudomonas* sp-SUK1 & *A. Ochraceus* NICM -1146 under solid state fermentation was analysed. The mixture of textile dyes was prepared by adding various textile dyes (each 20mg/L). Rice Bran, an agricultural low cost adsorbent was used to remove

color for mixture of dyes and removal was found to be 62% by ADMI method. The adsorbed mixture of textile dyes was then degraded by a Consoritum-PA under solid state fermentation which was found more efficient than individual micro-organism. ADMI results showed 82% decolorization in 36hrs for the adsorbed mixture of dyes by consortium-PA (Kadam *et al*., 2013). In 2010 studies were carried out to test the efficiency of electrocoagulation/electroflotation in removing color from synthetic and real textile wastewater by using aluminium and iron electrodes in an external-loop airlift reactor of 20 L. Removal efficiency was found to be different for both reactive, disperse and synthetic mixture of dyes. Aluminium electrodes gave better results for disperse dyes whereas iron electrode for reactive dyes and mixed synthetic dyes. The optimum pH for the process was observed to be 7.5 (Balla *et al.*, 2010).

To degrade a mixture of six commercial dyes photochemically, the degradation process was assisted by electrochemical method under halogen lamp. The COD removal percentage for mixture of dyes was found to be 73%. The mixture of dyes followed Langmuir –Hinshelwood isotherm and pseudo first order kinetics (Zainal *et al.*, 2007). Textile wastewater was decolorized containing acid dyes in Upflow Anaerobic Sludge Blanket (UASB) reactor system under mixed anaerobic granular sludge. A mixture of three dyes was prepared and treated using UASB reactor system achieving maximum decolorization up to $89\pm1.86\%$ at 300 mg/l dye concentration. The COD removal reduced from $93\pm1.9\%$ to $85\pm2.3\%$ at 10 mg/l and 300 mg/l dye concentrations, respectively indicating no severe toxicity caused to microorganism (Somasiri *et al.*, 2006). Mg/Pd^{4+} system was analysed for efficiency of removal of 80% to 95% color removal was obtained within 24 hours. Bimetallic system was found efficient in decolorizing mixture of commercial dyes (Patel and Suresh, 2006). In addition to this a combined Al (III) coagulation/activated carbon adsorption method was used for removal of reactive dyes from wastewater in two step method. Effects of various factors like pH, coagulant dosage, contact time and adsorbent dosage on dye removal were also studied. Coagulation technique was followed by adsorption which achieved almost the total elimination of dyes from wastewater, with 90% reduction in COD, TOC and adsorbable organic halide (AOX) (Papic *et al.*, 2004).

CONCLUSION

Wastewater discharged by textile finishing industries has become an great environmental concern for the scientists because of the prevailing hazards in our ecosystem. The dissolved dyes present in the effluent are resistant to light, acids, bases and oxygen, as these are the desired properties which should be possessed by dyed material. It is therefore difficult to treat textile wastewater especially when it contains a matrix of dyes. Real effluents generally include more than one component and study of the possible interactions between different chromophores will be very useful for the treatment of real effluents. Effect of various parameters like pH, temperature, time, elution and regeneration were noted which will prove to be essential factor during designing of the treatment (Biati *et al.*, 2014). During investigation, data on decolorizing efficiencies of different methods, suitability of isotherm models, and kinetics of the method have also been mentioned. Use of biosorbents combined with other techniques would give better results and will be economically sound. Along with dye removal several treatment methods tried to reduce COD, BOD and increased the quality of treated water which may be further used after treatment. Although some techniques were able to fully decolorize the matrix of dyes but several other factors which interfere may also be considered. Besides high efficiency of dye removal, the combined treatment processes offer many advantages for potential application such as coagulant savings, minimal amount of sludge formation and also economic feasibility.

Due to the foreseen water scarcity wastewater treatment is the issue to be taken up on priority. An attempt has been made to review the efficiency of various techniques used for decolorization of mixture of dyes and the use of combined techniques for successful water management.

ABBREVIATIONS
ADMI = *American Dye Manufacture Institute*
ANOVA = *Analysis of Variance*

AOX = *Advanced Oxidation Processes*
Adsorbable Organic Halide
BOD = *Bichemical Oxygen Demand*
CNT-PTFE = *Carbon Nano Tube- Poly Tetra Fluoro Ethylene*
COD = *Chemical Oxygen Demand*
EPIDMA = *Epichlorohydrin Dimethyl amine*
FTIR = *Fourier Transform Infrared Spectroscopy*
GC-MS = *Gas Chromatography Mass Spectroscopy*
HOBT = *HydroxyBenzoTriazole*
HPLC = *High Performance Liquid Chromatography*
HPTLC = *High Performance Thin Layer Chromatography*
PDADMAC = *Poly Diallydimethyl Ammonium Chloride*
PEA = *Photo Electro Adsorption*
PEPIDA = *Poly Epichlorohydrin Diamine*
PVA = *Phosphorylated Polyvinyl Alcohol*
RBC = *Rotating Biological Contactor Reactor*
RTE = *Real Textile Effluent*
TOC = *Total Organic Carbon*
UASB = *Upflow Anaerobic Sludge Blanket*

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