Review article

Removal of synthetic dyes from wastewaters: a review

Esther Forgacs[a,*], Tibor Cserháti[a], Gyula Oros[b]

[a] Research Laboratory of Materials and Environmental Chemistry, Chemical Research Center, Hungarian Academy of Sciences,
P.O. Box 17, 1525 Budapest, Hungary
[b] Plant Protection Institute, Hungarian Academy of Sciences, Pf. 102, Budapest 114, 1525, Hungary

Abstract

The more recent methods for the removal of synthetic dyes from waters and wastewater are complied. The various methods of removal such as adsorption on various sorbents, chemical decomposition by oxidation, photodegradation, and microbiological decoloration, employing activated sludge, pure cultures and microbe consortiums are described. The advantages and disadvantages of the various methods are discussed and their efficacies are compared.
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1. Introduction

Synthetic dyes are extensively used in many fields of up-to-date technology, e.g., in various branches of the textile industry (Gupta et al., 1992; Shukla and Gupta, 1992; Sokolowska-Gajda et al., 1996), of the leather tanning industry (Tünay et al., 1999; Kabadasil et al., 1999) in paper production (Ivanov et al., 1996), in food technology (Bhat and Mathur, 1998; Slampova et al., 2001), in agricultural research (Cook and Linden, 1997; Kross et al., 1996), in light-harvesting arrays (Wagner and Lindsey, 1996), in photoelectrochemical cells (Wrobel et al., 2001), and in hair colorings (Scarpi et al., 1998). Moreover, synthetic dyes have been employed for the control of the efficacy of sewage (Morgan-Sagastume et al., 1997) and wastewater treatment (Hsu and Chiang, 1997; Orhon et al., 1999), for the determination of specific surface area of activated sludge (Sorensen and Wakeman, 1996) for ground water tracing (Field et al., 1995), etc.

Synthetic dyes exhibit considerable structural diversity (Fig. 1). The chemical classes of dyes employed more frequently on industrial scale are the azo, anthraquinone, sulfur, indigoid, triphenylmethyl (trityl), and phthalocyanine derivatives. However, it has to be emphasized that the overwhelming majority of synthetic dyes currently used in the industry are azo derivatives. It should be noted that azo-keto hydrazone equilibria can be a vital factor in the easy breakdown of many of the azo dye system. Some dyes quoted in the review have only a marginal importance from the point of view of industrial application.

Unfortunately, the exact amount of dyes produced in the world is not known. It is estimated to be over 10,000 tons per year. Exact data on the quantity of dyes discharged in the environment are also not available. It is assumed that a loss of 1–2% in production and 1–10% loss in use are a fair estimate. For reactive dyes, this figure can be about 4%. Due to large-scale production and extensive application, synthetic dyes can cause considerable environmental pollution and are serious health-risk factors. Although, the growing impact of environmental protection on industrial development promotes the development of ecofriendly technologies (Desphande, 2001), reduced consumption of freshwater and lower output of wastewater (Knittel and Schollmeyer, 1996; Petek and Glavic, 1996), the release of important amounts of synthetic dyes to the environment causes public concern, legislation problems and are a serious challenge to environmental scientists.

Because of their commercial importance, the impact (Guaratini and Zanoni, 2000) and toxicity (Walthall and Stark, 1999; Tsuda et al., 2001) of dyes that are released in
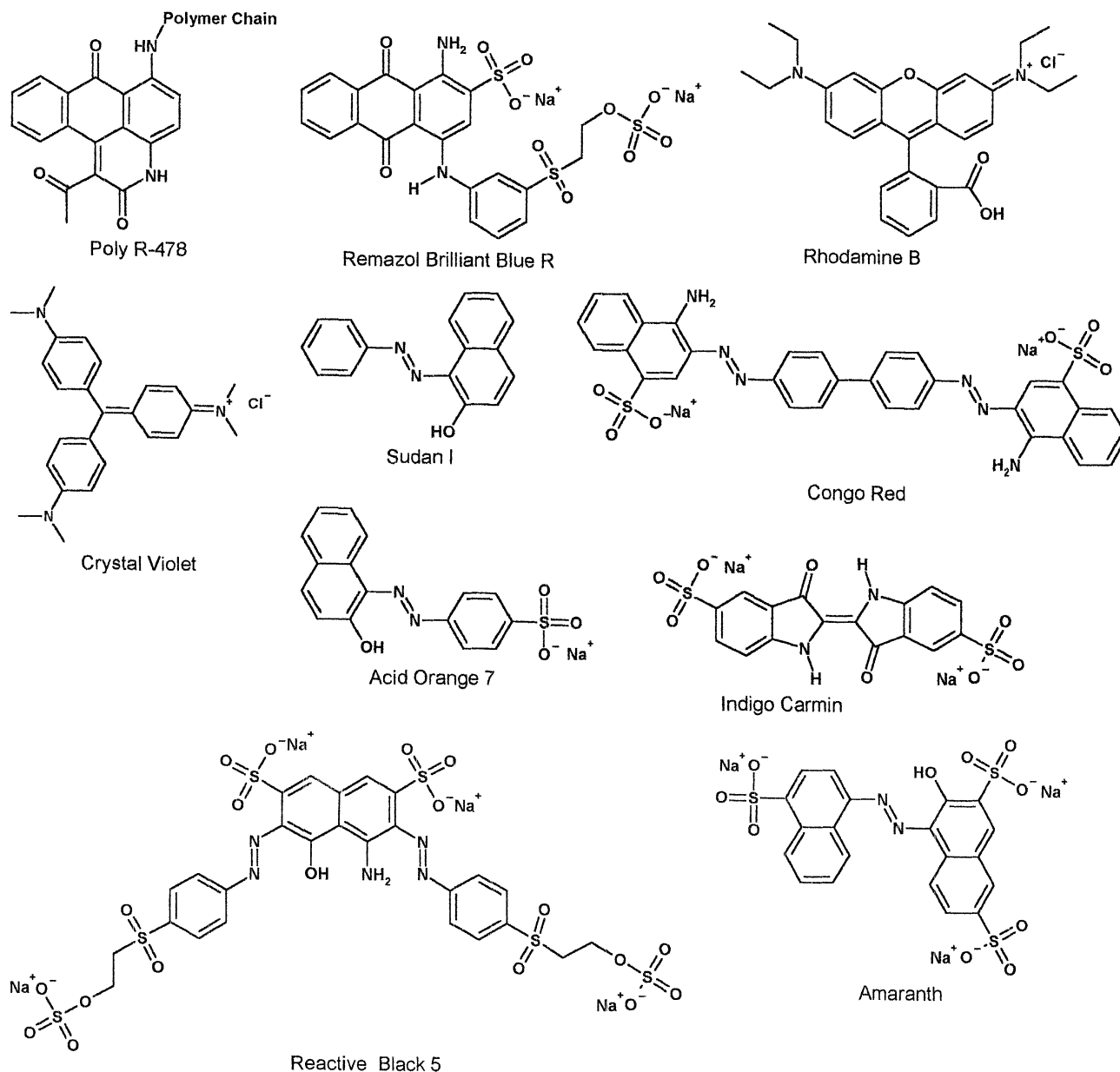
Fig. 1. The chemical structure of synthetic dyes most frequently studied in degradation experiments.

the environment have been extensively studied (Hunger, 1995; Calin and Miron, 1995). The formation of a carcinogenic amine from the dye Direct Blue 14 by human skin bacteria (Platzek et al., 1999) and the antifungal activity of 13 diazobenzene dyes have been established (Oros et al., 2001). As several thousand different synthetic dyes that are employed exhibit various biological activities, it is understandable that our knowledge concerning their behavior in the environment and health hazards involved in their use is still incomplete.

Traditional wastewater treatment technologies have proven to be markedly ineffective for handling wastewater of synthetic textile dyes because of the chemical stability of these pollutants. Thus, it has been verified that, of the

18 azo dyes studied 11 compounds passed through the activated sludge process practically untreated, 4 (Acid Blue 113, Acid Red 151, Direct Violet 9, and Direct Violet 28) were adsorbed on the waste activated sludge and only 3 (Acid Orange 7, Acid Orange 8, and Acid Red 88) were biodegraded (Shaul et al., 1991).

A wide range of methods has been developed for the removal of synthetic dyes from waters and wastewaters to decrease their impact on the environment. The technologies involve adsorption on inorganic or organic matrices, decolorization by photocatalysis, and/or by oxidation processes, microbiological or enzymatic decomposition, etc. (Hao et al., 2000). The efficacy of the various methods of dye removal, such as chemical precipitation, chemical oxidation,

adsorption along with their effects on subsequent biological treatment was compared in an earlier paper (Tunay et al., 1996). Chemical oxidation was very effective but the efficiency strongly influenced by the type of oxidant.

The objectives of this review are the compilation of the newer achievements in the technologies developed for the removal of synthetic dyes from water and wastewater, classification and short description of the methods, critical evaluation of the technological processes and the comparison of their advantages and disadvantages.

2. Removal of synthetic dyes from wastewaters by adsorption and other physicochemical methods

As synthetic dyes in wastewater cannot be efficiently decolorized by traditional methods, the adsorption of synthetic dyes on inexpensive and efficient solid supports was considered as a simple and economical method for their removal from water and wastewater. The adsorption characteristics of a wide variety of inorganic and organic supports have been measured and their capacity to remove synthetic dyes has been evaluated.

2.1. Inorganic supports

Because of their good mechanical and chemical stability, high specific surface area and resistance to microbiological degradation-specific inorganic supports have been preferentially applied in adsorption studies.

2.1.1. Carbon-based inorganic supports

The excellent adsorption properties of carbon-based supports have been exploited for the decolorization of dyes in the industrial effluents. For a better understanding of the physicochemistry of adsorption processes on the carbon surface, a homogeneous surface diffusion model was developed and successfully applied for the description of the adsorption of dyes and other wastes on the surface of granulated activated carbon (Roy et al., 1993).

It has been repeatedly shown that the type of carbon sorbent and its mode of preparation exert a marked influence on the adsorption capacity. It has been found that the adsorption characteristics of lignite-based carbon markedly depended on the mode of preparation. Maximal dye adsorption was achieved by a lignite that has been treated with 50% solution of sodium tungstate at 800 °C (Duggan and Allen, 1997). The efficacy of various wood charcoals has also been compared for the removal of Basic Red 22 and Acid Blue 25 from textile mill effluents. The data indicated that the media obtained from fluted charcoal, pine and chestnut trees were superior to lamellar charcoal, and to the media acquired from beech wood and oak trees (Marmier-Dussoubs et al., 1991). The adsorption process on the surface of carbonized spent bleaching earth has been studied in detail. Basic Blue 3, Methylene Blue, Acid Blue and

Reactive Yellow 2 were adsorbed on this sorbent. It has been demonstrated even in this instance that the mode of preparation of the sorbent had a considerable influence on the adsorption parameters, whereas the effect of pH was negligible (Low et al., 1995).

The good sorption characteristics of sulfonated coals for the removal of synthetic dyes have also been demonstrated (Mittal and Venkobachar, 1993). It has been further found that powdered active carbon efficiently removes the azo dyes Orange P and Red Px from wastewater (Danis et al., 1999). A similar study proved that granular activated carbon can bind acid dyes (Walker and Weatherley, 1997) and the kinetics of adsorption have been elucidated (Walker and Weatherley, 1999).

The results discussed above clearly state that carbon-based sorbents show excellent adsorption properties for a considerable number of synthetic dyes (Table 1). However, the preparation of carbon sorbents is generally energy consuming. Consequently, the commercially available products are fairly expensive. Since a large amount of carbon sorbent is needed for the removal of dyes from a large volume of effluent, the expenses involved hamper their application.

2.1.2. Other inorganic supports

As substituents for carbon-based sorbents the adsorption capacity of a wide variety of other inorganic supports was also measured using different dye-support pairs.

The efficacy of coal, fly ash, wollastonite, and china clay was compared for the removal of Omega Chrom Red ME from effluents. It was found that each sorbent could be employed in the adsorption process (Shukla and Gupta, 1992). The use of acid-activated clay for the removal of basic, acidic, disperse, direct and reactive dyes was also reported. The highest adsorption capacity was observed for basic dyes and the support was proposed as an efficient adsorption medium for their removal from aqueous solution (Juang et al., 1997). Furthermore, china clay was found to be an effective sorbent for the removal of Omega Chrom Red ME from house wastewater. An acidic pH, low temperature and smaller particle size of china clay increased the efficacy of removal (Gupta et al., 1992).

The good adsorption capacity of silica was exploited in the removal of the textile dye Basic Blue 3 from effluents (Ahmed and Ram, 1992) and was employed for the adsorption of Rhodamine B, Acid Red 4, and Nile Blue sulfate from aqueous solutions (Saleem et al., 1993). Alumina has also been used for the removal of Rhodamine B and Nile Blue Sulfate from wastewater (Salem et al., 1994). It has been established that the adsorption of dyes on alumina follows the Langmuir isotherm equation and the analysis of thermodynamical parameters revealed that the adsorption of these dyes is more favorable at high temperatures. Not only pure Al_2O_3 but also waste red mud, a by-product of aluminium production were employed for the removal of Congo Red from wastewater using, 90-min of equilibrium time (Namasivayam and Arasi, 1997).

Table 1
List of organisms intensively decolorizing synthetic dyes

No. of organism	Dye	Reference
Prokaryota		
Gram negative bacteria		
(1) *Aeromonas hydrophila*	Acid Orange 7, Acid Red 106, Direct Orange 39, Direct Yellow 4, Direct Yellow 12, Reactive Black NR, Reactive Blue 160, Reactive Blue 222, Reactive Red 198	Chen et al. (2003)
(2) *Burkholderia cepacia*	Acid Orange 7, Anthraquinone-2-sulfonate, Remazol Red F3B	Laszlo (2000)
(3) *Citrobacter* sp.	Brilliant Green, Crystal Violet, Gentian Violet, Malachite Green, Methyl Red,	An et al. (2002)
(4) *Desulfovibrio desulfuricans*	Reactive Orange 96	Yoo et al. (2001)
(5) *Escherichia coli*	Ethyl Red, Methyl Red	Nakanishi et al. (2001)
	Reactive Red 22	Chang and Lin (2001)
	Reactive Red 22	Chang and Kuo (2000)
(6) *Geobacter sulfurreducens*	Anthraquinone-2,6-disulfonate	Cervantes et al. (2003)
(7) *Klebsiella pneumoniae*	Methyl Red	Wong and Yuen (1998)
(8) *Proteus mirabilis*	Deep Red	Chen et al. (1999)
(9) *Pseudomonas luteola*	Crystal Violet, Red Pigment 2B, Red Pigment V2	Hu (1998, 2001)
	Reactive Red 22	Chang et al. (2001), Chen (2002)
(10) *Pseudomonas mendocina*	Methyl Violet	Sarnaik and Kanekar (1999)
(11) *Pseudomonas putida*	Tectilon Blue 4R-01	Walker and Weatherley (2000)
(12) *Pseudomonas stutzeri*	Methyl Red	Itoh et al. (2002)
(13) *Sphingomonas xenophaga*	Acid Orange 7, Acid Orange 8, Acid Orange 10, Acid Red 4, Acid Red 88	Coughlin et al. (1999)
	Congo Red	Diniz et al. (2002)
	Naphthalene-2-sulfonate, Naphtol Blue Black	Keck et al. (2002)
	Reactive Red 2	Zee van der et al. (2003)
(14) *Stenotrophomonas maltophilia*	Crystal Violet	Kim et al. (2002a,b)
(15) *Xenophilus azovorans*	Carboxy-Orange II	Blumel et al. (2002)

Table 1 (*continued*)

No. of organism	Dye	Reference
Gram positive bacteria		
(16) *Arthobacter globiformis*	Acridine Orange, Crystal Violet	Itoh et al. (1998a,b)
(17) *Bacillus benzenovorans*	Tectilon Blue 4R-01	Walker and Weatherley (2000)
(18) *Bacillus cereus*	Azobenzene	Koneva and Kruglov (2001)
(19) *Bacillus gordonae*	Tectilon Blue 4R-01	Walker and Weatherley (2000)
(20) *Bacillus polymixa*	Azobenzene	Koneva and Kruglov (2001)
(21) *Caulobacter subvibrioides*	Acid Orange 6, Acid Orange 7, Acid Orange 8, Acid Orange 12, Acid Red 151, Acid Red 88, Methyl Red	Mazumder et al. (1999)
(22) *Clostridium perfringens*	Amaranth	Semde et al. (1998)
	Bromophenol Blue, Crystal Violet, Methyl Orange	Kim et al. (2002a,b)
(23) *Kurthia* sp.	Brilliant Green, Crystal Violet, Magenta	Wong and Yuen (1998)
	Malachite Green	Sani and Banerjee (1999)
	Pararosaniline	Wong and Yuen (1998)
(24) *Nocardia corallina*	Crystal Violet	Azmi et al. (1998)
(25) *Paenibacillus azoreducens*	Remazol Black B	Meehan et al. (2001)
(26) *Streptomyces viridosporus*	Poly R-478	Ball and Colton (1996)
Eukaryota		
Yeasts—Ascomycota		
(27) *Candida curvata*	Chrysoidine	Kakuta et al. (1998)
(28) *Candida lipolytica*	Reactive Blue 19	Aksu and Donmez (2003)
(29) *Candida tropicalis*	Reactive Black 5, Reactive Blue 19, Reactive Red	Donmez (2002)
(30) *Candida zeylanoides*	Acid Orange 7	Ramalho et al. (2002)
	Azobenzenesulfonates	Martins et al. (1999)
	p-Methoxyazobenzene	Martins et al. (1999)
(31) *Geotrichum candidum*	Reactive Blue 5	Lee et al. (2000)
	Reactive Black 5, Reactive Red 158, Reactive Yellow 27	Maximo et al. (2003)
(32) *Kluyveromyces marxianus*	Reactive Blue 19	Aksu and Donmez (2003)
	Remazol Black B	Meehan et al. (2000), Bustard et al. (1998)
	Remazol Red	Bustard et al. (1998)
	Remazol Turquoise Blue	Bustard et al. (1998)

Table 1 (*continued*)

No. of organism	Dye	Reference
Eukaryota		
Yeasts—Ascomycota		
(33) *Pichia anomala*	Disperse Red 15	Itoh et al. (1996)
(34) *Saccharomyces cerevisiae*	Reactive Blue 19	Aksu and Donmez (2003)
(35) *Schizosaccharomyces pombe*	Reactive Blue 19	Aksu and Donmez (2003)
Filamentous Fungi—Ascomycota		
(36) *Aspergillus ficuum*	Direct Black 22	Dong et al. (2001)
(37) *Aspergillus foetidus*	Drimarene Black HFGR1	Sumathi and Manju (2000)
	Drimarene Navy BF Blue F2G1	Sumathi and Manju (2000)
	Drimarene Red BR F3B1	Sumathi and Manju (2000)
(38) *Aspergillus niger*	Congo Red	Fu and Viraraghavan (2002)
(39) *Myceliophthora thermophila*	Poly R-478	Alcade et al. (2002)
(40) *Penicillium* sp.	Poly R-478, Poly S-119	Zheng et al. (1999)
(41) *Rhizopus arrhizus*	Reactive Orange 16	O'Mahony et al. (2002)
Filamentous fungi—Basidiomycota		
(42) *Bjerkandera adusta*	Amaranth, Remazol Black B, Remazol Orange, Tropaeolin O	Swamy and Ramsay (1999a,b)
	Reactive Blue 15, Reactive Blue 38	Heinfling-Weidtmann et al. (2001)
(43) *Coprinus cinereus*	Direct Blue 1	Schneider et al. (1999)
(44) *Coriolus versicolor*	Acid Orange 7	Sam and Yesilada (2001); Lin et al. (2003)
	Everzol Turquoise Blue G	Kapdas and Kargi (2002)
	Methylene Blue	Mazmanci et al. (2002)
	Pigment Violet 12	Itoh et al. (1998a,b)
(45) *Cunninghamella elegans*	Malachite Green	Cha et al. (2001)
(46) *Cunninghamella polymorpha*	Disperse Blue 60	Sugimori et al. (1999)
(47) *Datronica concentrica*	Poly R-478	Tekere et al. (2001)
(48) *Dichotmius sqaluens*	Brilliant Green, Cresol Red, Crystal Violet	Gill et al. (2002)
(49) *Eichhornia crassipes*	Acid Blue 25	Lee et al. (1999)
	Reactive Blue 2	Lee et al. (1999)
(50) *Flavodon flavus*	Azure B, Congo Red, Poly B 411, Reactive Blue 19	Raghukumar (2000)
	Congo Red	Tatarko and Bumpus (1998)
(51) *Funalia trogii*	Acid Orange 7	Sam and Yesilada (2001)
(52) *Ganoderma* sp.	Reactive Blue 19	Maximo et al. (2003)
(53) *Irpex lacteus*	Reactive Blue 19	Maximo et al. (2003), Bhatt et al. (2000), Kasinath et al. (2003)

Table 1 (*continued*)

No. of organism	Dye	Reference
Filamentous fungi—Basidiomycota		
(54) *Lentinula edodes*	Amido Black 10 B, Bromophenol Blue, Methyl Red, Reactive Blue 19	Nagai et al. (2002)
	Acid Orange 7	Hatvani and Mecs (2002)
	Poly R-478	Chiu et al. (1999), Hatvani and Mecs (2002)
(55) *Phanerochaete chrysosporium*	Amaranth	Swamy and Ramsay (1999a,b)
	Disperse Orange K-GL, Everzol Yellow 4GL, Everzol Red RBN, Everdirect Supra yellow PG, Everzol Turquoise Blue G	Kapdan et al. (2000)
	Indigo Carmine	Podgornik et al. (2001), Gemeay et al. (2003)
	Poly R-478	Couto et al. (2000a), Mielgo et al. (2002)
	Red HE 8B	Sani et al. (1998)
	Remazol Black B	Swamy and Ramsay (1999a,b)
	Remazol Turquoise Blue	Conneely et al. (1999)
(56) *Phanerochaete magnoliae*	Reactive Blue 19, Reactive Red 158, Reactive Yellow 27	Maximo et al. (2003)
Phanerochete crassa	Poly R-478	Takano et al. (2001)
(57) *Phellinus gilvus*	Indigo	Balan and Monteiro (2001)
(58) *Phlebia fascicularia*	Brilliant Green, Cresol Red, Crystal Violet	Gill et al. (2002)
(59) *Phlebia floridensis*	Brilliant Green, Cresol Red, Crystal Violet	Gill et al. (2002)
(60) *Phlebia tremellosa*	Remazol Black B	Kirby et al. (2000)
(61) *Pleurotus eryngii*	Reactive Black 5	Heinfling et al. (1998)
(62) *Pleurotus ostreatus*	Eosin Yellowish, Evans Blue Phenol, Red Poly B 411	Eichlerova et al. (2002)
(63) *Pleurotus pulmonarius*	Amido Black 10 B, Brilliant Cresyl Blue, Congo Red, Ethyl Violet, Methyl Green, Methyl Violet, Reactive Blue 19, Trypan Blue	Zilly et al. (2002)
(64) *Pleurotus sajor-caju*	Indigo	Balan and Monteiro (2001)
(65) *Pycnoporus cinnabarinus*	Reactive Blue 19	Balan and Monteiro (2001)
(66) *Pycnoporus sanguineus*	Bromophenol Blue	Pointing et al. (2000)
	Indigo	Balan and Monteiro (2001)
	Malachite Green	Pointing et al. (2000)

(*continued on next page*)

Table 1 (continued)

No. of organism	Dye	Reference
Filamentous fungi—Basidiomycota		
(67) *Rigidoporus* sp.	Reactive Blue 19, Reactive Red 158, Reactive Yellow 27	Maximo et al. (2003)
(68) *Sclerotium rolfsii*	Acid Blue 74, Reactive Blue 19	Nyanhongo et al. (2002)
(69) *Trametes cingulata*	Cresol Red, Poly R-478	Tekere et al. (2001)
(70) *Trametes hirsuta*	Acid Blue 225, Basic Red 9, Direct Blue 71	Nyanhongo et al. (2002)
	Crystal Violet	Abadulla et al. (2000)
	Indigo	Campos et al. (2001)
	Poly R-478	Maceiras et al. (2001)
	Reactive Blue 19, Reactive Blue 221	Nyanhongo et al. (2002)
(71) *Trametes modesta*	Acid Blue 74, Reactive Blue 221, Direct Blue 71, Basic Red 9, Acid Blue 225, Reactive Blue 19	Nyanhongo et al. (2002)
(72) *Trametes pocas*	Cresol Red, Crystal Violet	Tekere et al. (2001)
(73) *Trametes trogii*	Poly R-478	Lenin et al. (2002)
(74) *Trametes versicolo*	Acid Blue 74	Nyanhongo et al. (2002)
	Acid Violet 7	Zhang and Yu (2000)
	Acid Violet 17	Nyanhongo et al. (2002)
	Amaranth	Swamy and Ramsay (1999a,b), Ramsay and Nguyen (2002), Shin et al. (2002)
	Brilliant Blue R	Borchert and Libra (2001)
	Congo Red	Ramsay and Nguyen (2002)
	Phenol Red	Lorenzo et al. (2002)
	Poly R-478	Leidig et al. (1999)
	Ponceau Red 4R	Keharia and Madamwar (2002)
	Procion Red	Keharia and Madamwar (2002)
	Reactive Black 5	Maximo et al. (2003), Borchert and Libra (2001), Ramsay and Nguyen (2002)
	Reactive Blue 15	Ramsay and Nguyen (2002), Swamy and Ramsay (1999a,b)
	Reactive Blue 19	Borchert and Libra (2001), Minussi et al. (2001), Nyanhongo et al. (2002)
	Reactive Blue 28	Keharia and Madamwar (2002)
	Reactive Blue 221	Nyanhongo et al. (2002)
	Reactive Golden Yellow R	Keharia and Madamwar (2002)

Table 1 (continued)

No. of organism	Dye	Reference
Filamentous fungi—Basidiomycota		
	Reactive Red 158	Maximo et al. (2003)
	Reactive Red 198	Borchert and Libra (2001)
	Reactive Violet 5	Keharia and Madamwar (2002)
	Reactive Yellow 27	Maximo et al. (2003)
	Remazol Black B	Swamy and Ramsay (1999a,b)
	Remazol Orange	Swamy and Ramsay (1999a,b)
	Tropaeolin O	Swamy and Ramsay (1999a,b), Ramsay and Nguyen (2002)
(75) *Trametes villosa*	Reactive Blue 19	Minussi et al. (2001)
Algae		
(76) *Chlorella pyrenoidosa*	Direct Brown NM	Huang et al. (2000)
(77) *Spirogyra* sp.	Reactive Yellow 22	Blumel et al. (2002)
Planta		
(78) *Mentha puligeum*	Poly R-478	Strycharz and Shetty, 2002
(79) *Rosmarinus officinalis*	Poly S-119	Zheng et al. (1999)
(80) *Wolffia arrhiza*	Methyl Violet	Kanekar et al. (1993)
(81) *Thymus vulgaris*	Poly S-119	Zheng et al. (1999)
(82) *Spirodella polyrrhiza*	Methyl Violet	Kanekar et al. (1993)

Vermiculite extract solutions as coagulants and vermiculite as the adsorbent were used for the removal of Basic Blue from dye wastewaters. The results indicated that the efficacy of vermiculite as both coagulant and adsorbent was better than that of conventional coagulants and adsorbents (Choi and Cho, 1996).

Aqueous solutions of Acridine Orange, Alcian Blue 8GX, Alizarin Red, Azure A, Azure B, Brilliant Blue G, Brilliant Blue R, Congo Red, Cresyl Violet Acetate, Crystal Violet, Eosin B, Eosin Y, Eryhtrosin B, Ehidium Bromide, Giemsa Stain, Janus Green B, Methylene Blue, Neutral Red, Nigrosin, Orcein, Propidium Iodide, Rose Bengal, Safranine 0, Toluidine Blue 0, and Trypan Blue were successfully decolorized passing the solution through a column that was previously filled with Amberlite XAD-16. The efficacy of removal was higher for dyes with low molecular mass, with lower flowrate and smaller particle size of the resin. The adsorbed dyes were easily extracted by washing the column with methanol (Lunn et al., 1994). The decontaminated solutions showed no mutagenicity towards *Salmonella typhimurium* (Lunn and Sansone, 1991).

The application of various inexpensive industrial wastes has also been elucidated for adsorption. Thus, a waste containing trivalent iron and trivalent chromium hydroxide

was employed for the removal of an azo dye from wastewater. The removal efficiency of color was 91% at pH 3. The adsorption followed both the Langmuir isotherm and the Freundlich isotherm and was governed by ion exchange processes between the polar substructures of the azo dye and the hydrophilic adsorption centers of the sorbent (Namasivayam and Senthilkumar, 1995; Namasivayam et al., 1994). The aninoic dye New Coccine was also effectively adsorbed on sludge particles (Wang et al., 1998).

The findings compiled above suggest that carbon-based supports can be replaced by other, less expensive ones.

2.2. Organic supports

Organic supports have some advantages over inorganic ones. They generally originate from renewable sources and are wastes or by-products of industrial processes without any commercial value. The adsorption capacity of a number of such supports has been determined for synthetic dyes as has their potential of their practical application were evaluated.

Biogas waste slurry was dried, powdered and used for the extraction of Rhodamine B from the wastewater of a textile plant. It was found that adsorption followed the Freundlich isotherm and the efficiency increased at acidic pH (optimum pH 2.3) (Namasivayam and Yamuna, 1992). Efficient removal of the direct dye Brilliant Yellow from aqueous media with cross-linked chitosan fiber was also detected (Yoshida and Takemori, 1997). The application of orange peel (cellulosic waste) for the adsorption of Congo Red, Procion Orange and Rhodamine B has also been assessed. It was determined that the adsorption can be described by Langmuir and Freundlich isotherms and follows first-order kinetics. Acidic pH condition promoted adsorption while alkaline pH condition enhanced the desorption of dyes (Namasivayam et al., 1996).

The application of pasteurized wastewater solids was assessed for the adsorption of Methylene Blue. It has been established that elimination of the biological activity of the solids was a prerequisite for effective adsorption (Dobbs et al., 1995). Bagasse pith was tested as a sorbent for the adsorption of dyes. The adsorption process was described by a three resistance number mass transfer model (external mass transport, macropore and micropore diffusion) (Al Duri et al., 1990). Waste banana pith was used for the removal of Rhodamine B from aqueous solutions. The maximum efficacy (87%) was observed at pH 4. It has been concluded from the results that waste banana pith offers an economical resource for the removal of dyes from wastewaters (Namasivayam et al., 1993). The results demonstrated that the rate constants markedly depend on the type of sorbents, with the higher values being attained on cellulosic waste orange peel.

An interesting approach was to use of dead and pulverized macrofungus, *Fomitopsis carnea* as a sorbent for the basic dyes Orlamar Red BG, Orlamar Blue G, and Orlamar Red GTL. Pulverized fungus proved to be a good sorbent for the dyes, the adsorption in creased with increasing pH of the dye wastewater and followed first-order kinetics (Mittal and Gupta, 1996).A continuous flow study showed that the non-living roots of water hyacinth can efficiently adsorb Acid Blue 25 and Reactive Blue 2 (Lee et al., 1999).

The adsorption methods, independently of the inorganic or organic character of the supports have some drawbacks. Since adsorption processes are generally not selective, the other components of the wastewater can also be adsorbed by the support and the competition among the adsorbates can influence the dye binding capacity of supports in an unpredictable manner. Moreover, an adsorption process removes the synthetic dyes from wastewater by concentrating them on the surface retaining their structure practically unchanged. When the support is to be regenerated, the fate of the resulting concentrated solution of dyes presents a problem that is not satisfactorily solved. Even the mineralization of dyes on the surface of support cannot be achieved. Large-scale applications based on the adsorption process have to take into consideration the problems discussed above.

2.3. Other physicochemical methods

Adsorptive bubble separation techniques (ion flotation, solvent sublation and adsorbing colloid flotation) resulted in the efficient removal (99%) of Direct Blue from wastewater (Horng and Huang, 1993). The application of coagulation processes for the removal of dyes from wastewater has also been assessed. The efficiencies dependent on the type of flocculant and on the pH of the medium (Koprivanac et al., 1993). Electrocoagulation was used for the effective removal of Acilan Blue from the wastewater of an operating textile plant in a bipolar packed-bed electrochemical reactor (Ogutveren et al., 1992).

3. Photocatalytic decolorization and oxidation of synthetic dyes

Commercial dyes are designed to resist photodegradation, so the selection of optimal photocatalytic conditions for the decolorization of dyes requires considerable expertise. Because of the significant commercial and environmental interest the efficacy of a large number of catalysts and irradiation conditions has been established for the decolorization of various synthetic dyes.

3.1. Photocatalysis and oxidation with hydrogen peroxide

Hydrogen peroxide has been frequently applied to the decolorization of synthetic dyes in waters. Hydrogen peroxide can effectively decolorize dye wastewaters in the presence of Fe(II) sulfate, with the higher rates of decolorization at higher concentrations of the reagents (Kuo, 1992).

Iron (III) with hydrogen peroxide were successfully employed for the degradation of the dye intermediate anthraquinone-2-sulfonic acid sodium salt (Kiwi et al., 1993). Not only an iron catalyst but also a UV/H_2O_2 oxidation process has been used for the decolorization of Reactive Black 5 in textile wastewaters (Ince and Gonenc, 1997). The degradation of anthraquinone sulfonate dyes was facilitated by H_2O_2 in the presence of TiO_2 (Kiwi, 1994). Iron powder and hydrogen peroxide in combination were employed in the decolorization of Reactive Red 120, Direct Blue 160 and Acid Blue 40, in aqueous solutions. The non-biodegradable azo dye Orange II was effectively mineralized with iron and hydrogen peroxide, with pH exerting a considerable effect on the decomposition rate (Bandara et al., 1997). Optimal conditions for decolorization were found to be different for each dye, indicating that the development of a general oxidation method for a mixture of dyes would be very difficult. Thus, compromise must be made that is suitable for the decomposition of each dye at a reasonable oxidation rate (Tang and Chen, 1996). UV irradiation combined with hydrogen peroxide treatment was used for the decolorization of the mono-azo dyes Acid Red 1 and Acid Yellow 23. Decolorization followed a pseudo first-order rate profile and the rate increased with increasing concentration of H_2O_2 in the solution (Tang et al., 1997). Acetone as a photosensitizer has also been used to facilitate the photocatalytic degradation of Reactive Red 2 in aqueous solutions. The reaction followed pseudo first-order decay kinetics and involved both dechlorination and reduction (Tsui and Chu, 2001). The possibility of UV/H_2O_2 treatment of dyes with different structures has been studied in detail. The results indicated that the method could be successfully used for the decolorization of acid dyes, direct dyes, basic dyes and reactive dyes but it proved to be inadequate for vat dyes and disperse dyes (Yang et al., 1998).

3.2. Ozonation

Ozonation, as an effective oxidation process, has found application in the decolorization of synthetic dyes. The technique employed in the decoloration of Orange II. Oxalate. Formate and benzene sulfonate ions were the most important decomposition products (Tang and An, 1995a,b). It was reported that ozone effectively decomposed azo dyes in textile wastewater. The decomposition rate was considerably higher at acidic pH. However, the influence of temperature and UV irradiation on the decomposition rate was negligible (Koyuncu and Afsar, 1996). The negligible influence of UV irradiation on the decomposition rate of azo dyes by ozone has been supported by other authors. The effect of chemical structure on the decomposition rate has been demonstrated (Davis et al., 1994). The effect of ozonation on the toxicity of wastewater effluents has been investigated using the nematode *Caenorhabditis elegans*. The data indicated that the toxicity highly depended on the type of dye to be decomposed (Hitchcock et al., 1998). The

influence of operating parameters on the decolorization of a reactive dye by ozone has been studied in detail. The results indicated that the decomposition rate increased with increasing pH and temperature (Wu and Wang, 2001). A method employing a combination of membrane filtration with subsequent ozonation of retentates has been developed for the effective purification of colored textile wastewaters (Wu et al., 1998).

3.3. Photodecomposition in the presence of TiO_2

It has been proven that the presence of catalysts enhances the rate of photodecomposition. The role of TiO_2 in oxidation was studied. It was shown that the photodegradation rate of azo dyes under UV irradiation considerably depends on the chemical structure in the presence of TiO_2. Monoazo dyes were more easily decomposed than trisazo dyes, disazo dyes were not included in the experiment (Reutergardh and Iang-pashuk, 1997). UV irradiation and TiO_2 catalysis were used for the decomposition of Acid Blue 40. The initial step of photocatalytic decomposition was found to be hydroxyl radical attack to the carbon–nitrogen bond of the side chain of anthraquinone (Liakou et al., 1997a). The same system was applied for the study of the decomposition of Acid Blue 40, Basic Yellow 15, Direct Blue 87, Direct Blue 160 and Reactive Red 120. The data demonstrated that the oxidation mechanism was determined by both the pH and the chemical structure of the dyes (Liakou et al., 1997b). The efficacy of TiO_2 and cadmium sulfide (CdS) photocatalysts was compared in the photocatalytic decomposition of the Reactive Black 5. The initial pH had a different impact on CdS and TiO_2 photocatalysis, the first-order rate constant increased with increasing concentration of the semiconductor and with increasing light intensity and temperature. It was found that the toxicity of wastewater decreased with the presence of TiO_2 and increased with the presence of CdS (Shu et al., 1994). The azo dye Acid Orange 7 was successfully decomposed on titanium oxide particles in visible light in the presence of oxygen. Naphthoquinone and benzene sulfonic acid were identified as main decomposition products (Vinodgopal et al., 1996). Other dyes were also decolorized by TiO_2 and by irradiation in wastewater and it was found that the temperature did not influence markedly the decomposition rate of dyes (Shu and Huang, 1995).

3.4. Other oxidizing systems

The photodecomposition of five dyes (Reactive Red 2, Reactive Blue 4, Reactive Black 8, Basic Red 13 and Basic Yellow 2) under UV irradiation in the presence of trivalent iron-oxalato complexes was also reported (Nansheng et al., 1997a). It has been established that the rate of photodegradation is highly dependent on the chemical structure of the dye. The decomposition followed first-order kinetics. The acidic pH enhanced decomposition. The same dyes were subjected to photodegradation, using a trivalent iron-hy-

droxy catalyst and sunlight. The pseudo first-order decomposition rate was lower than under UV irradiation (Nansheng et al., 1997b). The photodegradation kinetics for five synthetic dyes, using Fe^{3+}-hydroxy and Fe^{3+}-oxalate complexes indicate that the rate constants were higher for Fe^{3+}-hydroxy complexes except for Reactive Red 2. This finding suggests that the Fe^{3+}-hydroxy system is more suitable for activation of the photodegradation of reactive dyes than is the Fe^{3+}-oxalate system. Optimal conditions for the chemical oxidation-decolorization process were determined with the help of modified Nernst model by using the NaOCl as oxidant by (Chang et al., 1996). Electrooxidation of Acid Blue 113 using a RuO_2/Ti electrode was reported to be successful (Mohan et al., 2001). It has been demonstrated that the azo dye Remazol Black B can be decomposed in an aqueous solution saturated with oxygen using a high-frequency ultrasonic generator (Vinodgopal et al., 1998).

4. Microbiological decomposition of synthetic dyes

The application of microorganisms for the biodegradation of synthetic dyes is an attractive and simole method by operation. However, the biological mechanisms can be complex. Large number of species has been tested (Table 1) for decoloration and mineralization of various dyes. Unfortunately, the majority of these compounds are chemically stable and resistant to microbiological attack. The isolation of new strains or the adaptation of existing ones to the decomposition of dyes will probably increase the efficacy of bioremediation of dyes in the near future.

The use of microorganisms for the removal of synthetic dyes from industrial effluents offers considerable advantages. The process is relatively inexpensive, the running costs are low and the end products of complete mineralization are not toxic. The various aspects of the microbiological decomposition of synthetic dyes have been previously reviewed by Stolz (2001). Besides the traditional wastewater cleaning technologies, other methods have been employed in the microbial decolorization of dyes. For instance, an activated sludge process was developed for the removal of Methyl violet and Rhodamine B from dyestuff effluents, using microorganisms that were derived from cattle dung (Kanekar and Sarnaik, 1991). Also in biofilms, efficient biodegradation of Acid Orange 7 has been demonstrated (Harmer and Bishop, 1992; Zhang et al., 1995). Azo dyes did not inhibit the capacity of biofilms in the removal of organics from wastewater (Fu et al., 1994). A multistage rotating biological contactor was used for the biodegradation of azo dyes, where an azo dye assimilating bacterium was immobilized in the system (Ogawa and Yatome, 1990).

4.1. Mixed cultures (microorganism consortiums)

The utilization of microbiotic consortiums offers considerable advantages over the use of pure cultures in the degradation of synthetic dyes. The individual strains may attack the dye molecule at different positions or may use decomposition products produced by another strain for further decomposition. However, it should be stressed that the composition of mixed cultures may change during the decomposition process, which interferes with the control of technologies using mixed cultures. Moreover, the efficacy of decomposition considerably depends on the chemical character of the synthetic dye and on the biodegradation capacity of the microorganism consortium. The benefits and drawbacks of the use of microbial consortiums for the decomposition and decolorization of various dyes have been previously reviewed by Banat et al. (1996). Optimal conditions for the microbial decolorization of dyes show marked diversity both in anaerobic and aerobic as well as mixed anaerobic/aerobic processes. However, it has been observed in a number of cases that the efficacy of aerobic treatment was inferior to that of anaerobic decolorization process.

4.1.1. Anaerobic decolorization of synthetic dyes

The efficacy of various anaerobic treatment applications for the degradation of a wide variety of synthetic dyes has been many times demonstrated (Delee et al., 1998). Experiments indicated that chemical reduction by sulphide is partially responsible for the anaerobic conversions of Acid Orange 7. Mathematical evaluation of the experimental results pointed out that autocatalysis played an important role where 1-amino-2-naphthol accelerated the chemical reduction of azo bond. (Zee van der et al., 2000). The decolorization of reactive water-soluble azo dyes was achieved under anaerobic conditions using glucose as a carbon source (Carliell et al., 1996). The supplement tapioca starch gave also enhanced the color removal efficacy from synthetic blue wastewater (Chinkewitvanich et al., 2000). Mordant Orange 1 and Azodisalicylate were reduced and decolorized under anaerobic conditions using methanogenic granular sludge (Razo-Flores et al., 1997). Reactive Red 141 was also decolorized under anaerobic conditions in a conventional sewage treatment technology. The chemical identification of the products of dye degradation showed that decolorization was via reduction mechanism (Carliell et al., 1994). The synthetic dye Tartrazine was found to be readily decolorized in an anaerobic baffled reactor (Bell et al., 2000; Plumb et al., 2001). Disperse Blue 79 was also reduced in anoxic sediment–water system, the primary decomposition products being N,N-disubstituted 1,4 azobenzene and 3-bromo-6-nitro-1,2-diaminobenzene (Weber and Adams, 1995). Great differences were observed among the decomposition rates of various dyes in anoxic settled bottom sediments. Half-life varied between a few days (Solvent Red 1) and some months (Solvent Yellow 33) (Baughman and Weber, 1994). The reactive azo dye Reactive Red 141 was decomposed under anaerobic conditions. The azo bonds were reduced and cleaved by the microbial community resulting in the

liberation of 2-aminonaphtalene-1,5 disulfonic acid (Carliell et al., 1995).

Much effort have been devoted to the study of the influence of various modern technologies on the decomposition rate of dyes and the effect of the presence of other compounds in the media. It has been recently established that the development of high-rate systems, in which hydraulic retention times are uncoupled from solids retention times, facilitate the removal of dyes from textile processing wastewaters (Lier van et al., 2001). Another study proved the feasibility of the application of anaerobic granular sludge for the total decolorization of 20 azo dyes (Zee van der et al., 2001a). It was further demonstrated that the application of the redox mediator anthraquinone-2,6-disulfonic acid highly accelerates the decomposition of azo dyes (Zee van der et al., 2001b). The effect of the presence of salts (nitrate and sulfate) on the decomposition rate of the azo dye Reactive Red 141 under anaerobic conditions has been studied. The results indicated that nitrate delays the onset of decomposition while sulfate did not influenced the biodegradation process (Carliell et al., 1998).

4.1.2. Anaerobic/aerobic decomposition of synthetic dyes

Although anaerobic reduction of azo dyes is generally more satisfactory than aerobic degradation, the intermediate products (carcinogenic aromatic amines) have to be degraded by an aerobic process. Diverse technologies have been developed for the successive anaerobic/aerobic treatment of dye wastewaters. It has been observed that the removal of dyes from wastewaters in an anaerobic–oxic system involved both decomposition by bacteria and adsorption onto the sludge. Decolorization rates were 20%, 72%, and 78% for Acid Yellow 17, Basic Blue 3, and Basic Red 2, respectively (An et al., 1996). This combined method has been successfully employed for the decomposition of bisazo vinylsulphonyl, anthraquinone vinylsulphonyl and anthraquinone monochlootriazine reactive dyes (Panswad and Luangdilok, 2000) and the considerable impact of the molecular structure on the decolorization rate has been demonstrated (Luangdilok and Panswad, 2000). Dye wastewaters were also treated using a sequential anaerobic/aerobic filter system. Results showed that, the Basic Red was removed very efficiently in the anaerobic filter, however, no removal of the Acid Yellow 17 occurred. (Basibuyuk and Forster, 1997). Another two-stage anaerobic/aerobic system successfully decomposed sulfonated azo dyes (Acid Orange 10, Acid Red 14, Acid Red 18) (FitzGerald and Bishop, 1995). It was further established that an anaerobic/aerobic treatment is suitable for the cleavage of the azo bond in various azo dyes (Seshadri et al., 1994). Moreover, it was found that biofilms degraded aerobically the azo dye Acid Orange 8. The azo bond of Acid Orange 8, Acid Orange 10 and Acid Red 14 was cleaved only under anaerobic conditions (Jiang and Bishop, 1994). The efficacy of the removal of reactive diazo Remazol Black B dye by aerobic and anoxic plus anaerobic/aerobic sequencing batch reactor

(SBR) activated sludge processes has been assessed. The results indicated that longer anoxic + anaerobic period promoted decolorization (Panswad et al., 2001). The azo dye Procion Red H-E7B has been efficiently decolorized in a combined anaerobic–aerobic process (O'Neill et al., 1999) and the beneficial effect of existence of carbohydrate at higher concentration on the decolorization has been proven (O'Neill et al., 2000).

4.2. Pure cultures of white-rot fungus

White-rot fungi produce a wide variety of extracellular enzymes (laccase, lignin peroxidase, phenol oxidase, Mn-dependent peroxidase and Mn-independent peroxidase) that decompose the highly stable natural (lignin, hemicellulose, cellulose, etc.). Because of their high biodegradation capacity they are of considerable biotechnological interest, and their application in the decolorization process of wastewaters has been extensively investigated (Young and Yu, 1997). Earlier results on decolorization of wastewaters by fungi have been reviewed (Fu and Viraraghavan, 2001).

4.2.1. Pure cultures of Phanerochaete chrysosporium

Because of its high enzyme production, the white rot fungus, *P. chrysosporium* has been frequently employed for the biodegradation of synthetic dyes. It was applied to the decoloration of Orange II, Tropaeolin 0, Congo Red and Azure B under aerobic conditions. The results indicated that the fungus can be used for the removal of these dyes from wastewater (Cripps et al., 1990). Decolorization was achieved in 6–9 days. Two strains of *P. chrysosporium* and an isolate of white-rot fungus were used for the decomposition of the azo dyes Amaranth, Orange G and the heterocyclic dye Azure B. The rate of decoloration of dyes depended on the composition of medium and on the dye-microorganism pair. It has been assumed that various extracellular peroxidases (lignin peroxidase and Mn-dependent peroxidase) or laccase are involved in the decolorization process. The data further indicated that the high decomposition rate of dyes can be achieved only by careful selection of the fungi and cultural conditions (Chao and Lee, 1994). The decomposition of Indigo Carmine by the fungus has also been studied and the involvement of ligninolytic enzymes in the process has been demonstrated (Podgornik et al., 2001). The biodegradation of Amaranth, New Coccine, Orange G and Tartrazine by *P. chrysosporium* and *Pleurotus sajor-caju* was compared. It was suggested that Mn-peroxidase, β-glucosidase and laccase can be involved in the decolorization process (Chagas and Durrant, 2001). It has been found that the addition of activators for the production of lignolytic enzymes by *P. chrysosporium* (Tween 80, veratryl alcohol, manganese (IV) oxide) increased the decomposition rate of the dye Poly R-478 (Couto et al., 2000a; Couto et al., 2000b). It has been confirmed that the lignin peroxidase of *P. chrysosporium*

removes not only dyes but also phenol and chlorophenol from wastewaters (Manamekalai and Swaminathan, 2000).

4.2.2. Pure cultures of other white-rot fungus

Other white-rot fungi have been used for the decoloration of different dyes. Thus, *Trametes versicolor* decomposed anthraquinone, azo and indigo-based dyes (Wang and Yu, 1998), *Pycnoporus cinnabarinus* rapidly decoloration Remazol Brilliant Blue in packed-bed bioreactor (Schliephaje and Lonergan, 1996), and *Trametes hirsuta* was able to decompose triarylmethane, indigoid and anthraquinone dyes (Abadulla et al., 2000). The degradation capacity of 103 strains of white-rot fungi has been measured. It has been established that the higher degradation rate was achieved by *Irpex lacteus* and *Pleurotus ostreatus* (Novotny et al., 2001). The biodegradation capacity of wood-rotting basidiomycete fungi was also determined using 14 structurally different synthetic dyes. The results indicated that the decomposition rate highly depends on both the chemical structure of the dye and the character of the fungi (Knapp et al., 1995). It has been determined that the presence of azoreductase in the microorganism and the permeation of the dye molecule are prerequisites of the microbial decolorization of dyes (Yatome et al., 1991a). The decoloration of the phthalocyanine dyes, Reactive Blue 15 and 38, by *Bjerkandera adusta* was studied in detail. It was found that the main metabolites were sulfophthalimides (Heinfling-Weidtmann et al., 2001). The potential of some white-rot fungi to decolorize indigo dye has been compared. The decomposition rate was the highest for *Phellinus gilvus* followed by those achieved *P. sajor-caju*, *Pycnoporus sanguineus* and *P. chrysosporium* (Balan and Monteiro, 2001). It has been reported that *Phlebia tremellosa* decomposes synthetic dyes but complete mineralisation did not occur (Kirby et al., 2000). The decoloration capacity of another set of white-rot fungi has been assessed using industrial dyes as model compounds. The results indicated that *Trametes hispida* produced lignolytic enzymes at higher rate than was achieved *Pleurotus ostreatus* in solid state cultures on whole oats (Rodriguez et al., 1999).

The assays carried out on another set of white rot fungi indicated that *Coriolus versicolor* showed the highest decomposition capacity (Knapp and Newby, 1999). An interesting combined method has been described for the decolorisation of Acid Violet 7. Pellets have been prepared form the mycelium of *T. versicolor* and activated carbon powder and their decoloration rate has been shown to be higher than those of the individual components (pure mycelium or activated carbon) (Zhang and Yu, 2000).

4.3. Other pure cultures

Although the capacity of white rot fungi to remove synthetic dyes from waters has been frequently demonstrated the search for other dye-decomposing microorganisms proceeds. Pure cultures other than white rot fungi have also found application in the decolorization of synthetic dyes.

The potential of fungi from marine habitats to degrade synthetic dyes (Azure B, Brilliant Green, Congo Red, Crystal Violet, Poly-R, Poly-B, Remazol Blue R) has been revealed (Raghukumar, 2000). Reactive azo dyes have been effectively removed from water by the fungus *Aspergillus foetidus*. However, the measurements indicated that the dyes were not decomposed, they were only adsorbed in the fungal biomass (Sumathi and Manju, 2000).

The ability of a *Kurthia* sp. to decolorize Magenta, Crystal Violet, Malachite Green, Pararosaniline and Brilliant Green has been reported (Sani and Banerjee, 1999). Pure culture of *Bacillus subtilis* can degrade *p*-aminoazobenzene under anoxic conditions, producing aniline and *p*-phenylenediamine as main decomposition products (Zissi and Lyberatos, 1996). The biodegradation of anthraquinone dyes by *B. subtilis* in industrial wastewater was also observed. The first step of biodegradation was the reduction of dyes to the leuko form (Itoh et al., 1993). It was reported that *B. subtilis* could decompose the triphenylmethane dye Crystal Violet at low concentrations below 7.10^{-6} mol/l, while *Escherichia coli* was ineffective. The main decomposition product was identified as 4,4′ -bis(dimethylamino) benzophenone (Yatome et al., 1991b). The capacity of *Pseudomonas* strains for the decolorization of various dyes has also been studied in detail (Yu et al., 2001). The azo dyes (Acid Violet 7, Acid Red 151 and Reactive Black 5) were degraded at higher extent (>90%) than Indigo Carmin, Acid Red 183 (chromium complex) and antraquinones (Reactive Blue 2 and Acid Green 27). It has been demonstrated that *P. mendocina* could effectively decolorize Methyl Violet in textile wastewater, with the use of a fixed-film reactor (Kanekar and Sarnaik, 1995; Kanekar et al., 1996). Decolorization of wastewaters containing reactive azo dyes was achieved by a culture of the bacteria *P. luteola*. Bacteria reduced the azo bond in Red G and biodegraded the other dyes (Hu, 1994). The capacity of *Klebsiella pneunomoniae* to decolorize Methyl Red under aerobic conditions was compared with that of *Acetobacter liquefaciens* in another study and the higher activity of *K. pneunomoniae* has been demonstrated (Wong and Yuen, 1996). It has been found that the sulfate reducing bacteria *Desulfovibrio desulfuricans* can also decolorize Reactive Orange 96 and Reactive Red 120 under anaerobic conditions (Yoo et al., 2000).

Food-borne bacteria are also capable for the reduction of dyes. The reduction of seven redox dyes by 13 food spoilage bacterial strains was determined. The results clearly show that the rate of reduction markedly varies among dye/organism pairs, proving the different reduction capacity of bacteria and the different sensitivity of dyes to reductases (Learoyd et al., 1992).

The biodegradation of azo dyes by the algae (*Chlorella pyrenoidosa*, *C. vulgaris* and *Oscillatoria tenuis*) has been also assessed. According to the data, the azo reductase of the algae is responsible for degrading azo dyes into aromatic amines by breaking the azo linkage. In addition, the algae

can play a direct role in degradation of azo dyes (Liu and Liu, 1992).

The application of microorganisms for the biodegradation of synthetic dyes is an attractive and simple method. Unfortunately, the majority of dyes are chemically stable and resistant to microbiological attack. The isolation of new strains or the adaptation of existing ones to the decomposition of dyes will probably increase the efficacy of microbiological degradation of dyes in the near future.

5. Enzymatic decomposition of synthetic dyes

The character of enzymes and enzyme systems in microorganisms that are suitable for the decomposition of dyes has been extensively investigated. Effort has been devoted to the separation, isolation and testing of these enzymes. Exact knowledge of the enzymatic processes governing the decomposition of dyes is important in the environmental protection both from theoretical and practical points of view.

Lignin peroxidase isoenzymes were isolated from *P. chrysosporium* and purified by chromatofocusing. The activity of isoenzymes towards decoloring triphenylmethane dyes, heterocyclic dyes, azo dyes and polymer dyes was compared with that of a crude enzyme preparation. Optimum pH values for the decolorization of dyes by various isozymes were markedly different. According to the results, the decomposition capacity of crude enzyme preparation

and purified isoenzymes showed marked differences while variations in the structure of dyes exerted slight influence (Ollikka et al., 1993). Horseradish peroxidase has been successfully employed for the decomposition and the precipitation of azo dyes. The degradation rate was dependent on the pH (Bhunia et al., 2001). Another study revealed that the enzymes of white rot fungus degraded Crystal Violet via *N*-demethylation (Bumpus et al., 1991). Interestingly, lignin peroxidase from *B. adusta* showed very low degradation capacity towards azo dyes and phthalocyanine dyes. However, veratryl alcohol considerably increased the decomposition rate (Heinfling et al., 1998). Similar investigations proved that pure laccase was also unable to decolorize Remazol Brilliant Blue R but the decoloration rate was facilitated by the presence of a mediator (violuric acid) (Soares et al., 2001).

The employment of enzyme preparations shows considerable benefits over the direct use of microorganisms. Commercial enzyme preparations can be easily standardized, facilitating accurate dosage. The application is simple and can be rapidly modified according to the character of the dye or dyes to be removed.

6. Future trends

The overwhelming majority of the current publications in the field of the removal of synthetic dyes from waters has

Table 2
Improvement of decolorization activity of organisms by interspecific transfer of genetic elements

Organisms		Function	References
Donor	Acceptor		
Prokaryotes			
Clostridium perfringens	*Escherichia coli*	Azoreductase	Rafii and Coleman (1999)
Bacillus sp.	*E. coli*	Azoreductase	Suzuki et al. (2001)
Rhodococcus sp.	*E. coli*	Azoreductase	Chang and Lin (2001)
Caulobacter subvibrioides	*E. coli*	Azoreductase	Govind et al. (1993)
Xenophilus azovorans	*E. coli*	Azoreductase	Blumel et al. (2002)
Pseudomonas luteola	*E. coli*	Azoreductase	Chang et al. (2000)
E. coli	*Sphingomonas xenophaga*	Flavin reductase	Russ et al. (2000)
Agrobacterium rhizogenes	*Mentha puligeum*	Tolerance to R-478	Strycharz and Shetty (2002)
Eukaryotes			
Geotrichum candidum	*Aspergillus oryzae*	Peroxidase	Sugano et al. (2000)
Ceriporiopsis subvermispora	*A. nidulans*	Peroxidase	Larrondo et al. (2001)
C. subvermisopra	*A. oryzae*	Peroxidase	Larrondo et al. (2001)
Coprinus cinereus	*Saccharomyces cerevisiae*	Laccase	Cherry et al. (1999)
C. cinereus	*A. oryzae*	Laccase	Schneider et al. (1999)
Coriolus versicolor	*Nicotiana tabacum*	Peroxidase	Iimura et al. (2002)
Phanerochaete chrysosporium	*A. nidulans*	Peroxidase	Larrondo et al. (2001)
P. chrysosporium	*A. oryzae*	Peroxidase	Larrondo et al. (2001)
Pycnoporus cinnabarinus	*Pychia pastoris*	Laccase	Otterbein et al. (2000)
P. cinnabarinus	*A. niger*	Laccase	Record et al. (2002)
Pleurotus sajor-caju	*P. pastoris*	Laccase	Soden et al. (2002)
Trametes versicolor	*S. cerevisiae*	Laccase	Larsson et al. (2001)
T. versicolor	*P. pastoris*	Laccase	O'Callaghan et al. (2002)
T. versicolor	*P. pastoris*	Laccase	Hong et al. (2002)
Armoracia rusticana	*S. cerevisiae*	Peroxidase	Morawski et al. (2001)

been dealing with the various aspects of the application of microbiological methods and techniques, with the search for new microorganisms providing higher decomposition rates and with the elucidation of the principal biochemical and biophysical processes underlying the decolorization of dyes. This trend unambiguously proves the decisive role of microbiological processes in the future technologies used for the removal of dyes from waters.

The widespread application of combined techniques using microbiological decomposition and chemical or physical treatments to enhance the efficacy of the microbiological decomposition can be expected in future.

Some new results indicate that gene manipulation; the creation of recombinant strains with higher biodegradation capacity will be applied in the future (Table 2). The cloning and expression in *E. coli* of an 'azoreductase' gene from *Clostridium perfringens* (Rafii and Coleman, 1999), from a *Bacillus* sp. (Suzuki et al., 2001), from *Pseudomonas luteola* (Hu, 1994) have been reported. Furthermore, the feasibility of the use of a recombinant *E. coli* strain, harboring azo-dye-decolorizing determinants from *Rhodococcus* sp. (Chang and Lin, 2001), and recombinant *Sphingomonas* sp. (Russ et al., 2000) for the decolorization of dye wastewater has been demonstrated. The exoenzymes of white-rot fungi have also been objects of genetic engineering. The laccase of various filamentous fungi was successfully transmitted into yeast. These manipulations enhanced the capacity of microorganisms to decolorize synthetic dyes. The expression of oxidases from higher plants augmented the catabolic potential of microbes (Haudenschild et al., 2000; Morawski et al., 2001) and in turn microbial genes straightened the tolerance of higher plant to Poly R-487 (Strycharz and Shetty, 2002; Iimura et al., 2002). Polymeric dye-tolerant plants may be useful in phytoremediation because they could provide a rhizosphere that was suitable for colonization by microbes that are efficient degraders of aromatic structures. The plant derived compounds can induce production of fungal redox enzymes (Curreli et al., 2001). Reductive cleavage of the azo bond dissipates the electron deficiency of the aromatic nuclei so that the aromatic amino compounds generated may be subject to subsequent oxidation and mineralization. The C-hydroxylation of aromatic rings by mammalian monoxygenases facilitates subsequent microbial degradation. Human cytochrome P450 enzymes are now routinely expressed as recombinant proteins in many different systems (Gillam, 1998; Sakaki and Inouye, 2000). The capacity of such recombinants to catabolize dyes has been tested (Stiborova et al., 2002). It is clear that complexity of association involved in the complete degradation should be increased with increasing complexity of the chemical structure of synthetic dyes. The genetically engineered microorganisms can accomplish degradation of synthetic dyes, which persist under normal natural conditions. In natural habitats, complex microbial/macrobial communities carry out biodegradation. Within them, a single organism may interact through interspecific transfer of metabolites. This co-metabolic potential may be complementary so that extensive biodegradation or even mineralization of xenobiotics can occur (Rieger et al., 2002). In this respect, deterioration of dyestuff effluents in constructed wetlands with multisite catabolic potential is a promising possibility. Mobilizing specific genes, encoding nonspecific multifunctional degradative sequences, may decisively increase the degradative potential of natural synthropic community against synthetic dyes. The use of recombinants that harbor dye-decolorizing determinants from other species can essentially enhance the capacity of waste remediation technologies.

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