Textiles

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ABSTRACT: A review of the literature published in 2008 on topics relating to wastewater treatment issues and technologies in the textile industries is presented. After a brief overview of the BATTLE project, the review is divided into the following sections: physico-chemical, biological and combined processes.

KEYWORDS: bacterial, color, decolorization, degradation, dyes, textiles.

BATTLE Project

The BATTLE project (Best Available Techniques for Water Reuse in Textile SMEs) is a LIFE funded project (LIFE05 ENV/IT/000846) which aims at demonstrating the technical–economical feasibility of a water recycling technique for textile companies. The main objective is the development and prototypal application of a clean technology for water reuse, to be proposed as

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reference BAT for small-medium (SMEs) size as well as for large enterprises.

Specific targets are therefore (Casarci et al., 2008):

– Evaluation of the BATs applicability for the SMEs of the textile finishing sector, as reported in the BREF (Best available techniques REFerence) document elaborated by the IPPC bureau (Directive 96/91/CE);

– Raising awareness on benefits and opportunities for the European SMEs and decision makers of textile and side sectors from the implementation of the new BATs.

According to Mattioli et al. (2008), when considering textile process effluents for possible reuse at viable technical economical conditions, a thorough segregation (wastewater design) is required. The study demonstrated that, despite the low concentration of most contaminants in the segregated effluents, they were not suitable for direct reuse mainly due to unacceptable levels of suspended solids and turbidity. Ultrafiltration (UF) was shown to be a feasible solution for effluents reclamation allowing an almost complete removal of the undesirable suspended material and colloids and a reduction of colour. Effluent reuse in textile production processes proved to be feasible, with exception of few textile operations. Operational data gained in the experimental activity were

used to design demonstrative scale treatment and reuse facilities and to make economical feasibility assessment.

Physico-Chemical Processes

Photodegradation. Baran et al. (2008) investigated the influence of various parameters such as the initial concentration of dye and catalyst (two types of TiO_2), the volume of irradiated solution and its absorbance on the kinetics of photocatalytic degradation of solutions containing individual azo dyes (Acid Black 1, Acid Orange 7, Basic Orange 66) and their mixtures. The results indicated that dyes photodegradation followed a pseudo first-order kinetic according to the Langmuir-Hinshelwood's heterogeneous catalysis model but only in the limited range of initial dyes concentrations. The photocatalytic process occurred only in a thin surface layer of the irradiated solution.

A review of the heterogeneous photocatalytic treatment of organic dyes in air and water was provided by Rajeshwar et al. (2008), including representative studies spanning approximately three decades. The review highlights that titanium oxide (TiO_2) was the most used inorganic semiconductor photocatalyst; other semiconductors such as ZnO, CdS, WO_3 and Fe_2O_3 were also used, albeit to a much smaller extent. Photocatalysis appears to be unsurpassed in its capability to remove color quickly (unlike biological treatment) and is known to be energy-efficient relative to Advanced Oxidation Process (AOP) competitors such as UV/H_2O_2. Other AOP candidates such as ozonation or UV/O_3 (and variants thereof), which are also very effective for dye decolorization and degradation, have accompanying disadvantages. On the technological front, the scientific advances must be translated to large-scale systems capable of continuously and reliably treating large volumes of dyeing wastewaters.

Rose Bengal (Acid Red 94 as Color Index, C.I.) was irradiated with UV light in the presence pf hydrogen peroxide and the photoinduced decolorization monitored spectrophotometrically. Dye decolorization increased with increasing H_2O_2 concentration and decreasing dye concentration. The maximum dye decolorization was determined as 90% with 0.005 mM dye at optimum 0.042 M H_2O_2 and pH 6.6. Additionally, it was seen that sulphite caused a maximum effect on percentage decolorization. The experimental data were also optimized using the response surface methodology (RSM) and the ANOVA results were evaluated (Rauf et al., 2008). Highly dispersed SiO_2/TiO_2 spherical core-shell particles were successfully developed by Lee et al. (2008) for the phocatalysis treatment of waste containing Methyl Orange (MO). Better dispersion of the particles was achieved when ultrasonication was applied during washing. The silica core crystals exhibited highly amorphous structures, whereas titania covered silica cores demonstrated pure anatase phase that grew in size upon heat treatment at 900°C. Complete decomposition of MO was achieved in less than 370 min using core-shell particles prepared from triple coating.

Hydrated titania was prepared by a sol-gel method, taking tetraisopropyl orthotitanate as starting material, and then promoted with different weight

percentage of sulfate by an incipient wetness impregnation method. The degradation of Methyl Orange (MO) under solar radiation was investigated to evaluate the photocatalytic activity of these materials. At 2.5 wt% sulfate loading, the average percentage of degradation of MO was nearly two times than that of neat TiO_2. The photocatalytic degradation followed first-order kinetics (Parida et al., 2008). Jain and Shrivastava (2008) investigated the photocatalytic degradation of a hazardous water soluble xanthene dye cyanosine in aqueous suspensions of titanium dioxide under a variety of conditions. They found optimum conditions for dye degradation to be: dye concentration of 1×10^{-4} M, pH 8, catalyst concentration of 0.04 g/L and temperature about 30°C. Photocatalytic degradation was also applied by Aleboyeh et al. (2008) to treat C.I. Acid Orange II in aqueous solution, using combined UV and hydrogen peroxide. By applying 3.75 W/L of the UV dosage in the reaction mixture of 17.5 mg/L dye and 525 mg/L H_2O_2, nearly 90% Total Organic Carbon (TOC) could be removed in 100 min.

Co-ZSM-5 catalysts with different Co-loadings (2-30 weight%) were prepared by El-Bahy et al. (2008) by using the incipient wetness impregnation method, and then characterized thoroughly. The assessment of the catalytic activity was performed by the use in the photodegradation of Acid Green (AG) dye as a probe reaction in presence of H_2O_2 as oxidant. A gradual increase of AG degradation rate was observed with increasing pH value and the optimum H_2O_2 concentration was 61.6 mmol/L. The experimental data pointed out the importance of both the cobalt moieties

and the zeolite framework structure in the AG degradation reaction. Körbahti and Rauf (2008a) investigated the decolorization of Toludine Blue (TB) in the presence of UV radiation and heterogeneous hybrid catalyst, namely titanium oxide impregnated with vanadium oxide. The process was optimized by using the Response Surface Methodology (RSM) and the optimum removal conditions were found to be: 26.5 mg/20 mL V_2O_5/TiO_2 catalyst concentration at pH 7.7. Under these conditions the complete decolorization was obtained at 50 μM TB concentration. The same authors (Körbahti and Rauf, 2008b) investigated also the photocatalytic degradation of Basic Red 2 (BR2) via UV radiation in the presence of H_2O_2 and optimized the process using the RSM. The optimized conditions were found to be: 20 μM BR2, 1.67 mM H_2O_2 and pH 7.6. In their study, Sohrabi and Ghavami (2008) tested the effects of different cations and C_2H_5OH and types of catalysts on the photocatalytic degradation of Direct Red 23 (scarlet F-4BS). After 90 min reaction, the relative decomposition order established was $UV/TiO_2 > UV/SnO_2 > UV/Fe_2O_3 > UV/ZnO$. The electrochemical-assisted photocatalytic process was successfully applied by Neelavannan and Basha (2008) for the degradation of textile washwater containing Procion Blue dye. After 7 h treatment, 90% COD and complete color removal were attained. The photodegradation of Methyl Orange (MO) was investigated by Chen et al. (2008) in aqueous suspension containing Ag/ZnO catalysts, successfully synthesized using a simple solvothermal method. MO could be completely mineralized after 60 min illumination.

Degradation of Congo Red (CR) was performed by means of photocatalysis of TiO_2 which was hydrothermally synthesized at 200°C in 2 h, in anatase phase with 8 nm crystallite size. Complete decolorization was achieved within 30 min irradiation (Erdemoğlu et al., 2008). Chatterjee et al. (2008) observed 90-95% decolorisation of triazine (Reactive Red 11, Reactive Red 2, and Reactive Orange 84) and vinylsulfone type (Reactive Orange 16 and Reactive Black 5) of reactive dyes upon prolonged illumination of the reacting system with 150 W xenon lamp at ambient conditions. According to Ullah and Dutta (2008), photodegradation efficiency of Methylene Blue (MB) with $ZnO:Mn^{2+}$ was significantly higher than that with undoped ZnO under visible light. The photocatalyst was also characterized with transmission electron microscopy (TEM), infrared spectroscopy (IS), photo-co-relation spectroscopy (PCS) and UV-vis-spectroscopy.

Advanced Oxidation Process. Tekbaş et al. (2008) investigated the heterogeneous photo-Fenton-type oxidation of the reactive azo dye Remazol Brilliant Orange 3R (C.I. Reactive Orange 16, RO16) solutions in a quartz batch reactor using artificial UVA as a light source and Fe-exchanged zeolite as a catalyst. The optimal operational parameters were found as follows: 35°C, pH as solution pH 5.2, 15 mmol H_2O_2 dosage, 1 g/L catalyst loading. The process achieved more than 90% decolorization of RO16 in 60 min; furthermore, it was possible to reuse the catalyst up to three successive experiments.

According to Hammami et al. (2008), the electro-Fenton process with the use of Pt or boron-doped diamond (BDD) anode and a carbon felt cathode was a very effective method for the degradation of Acid Orange 7 (AO7) azo dye in acidic aqueous medium of pH 3.0. Although the degradation/mineralization rate was higher with BDD anode at the beginning of the treatment, a mineralization ratio of 98% in terms of TOC removal was obtained after 9 h of electrolysis with both anodes. The absolute rate constant for the reaction between AO7 and •OH was determined as $(1.10\pm0.04)\times1010$ $M^{-1}s^{-1}$ from the competition kinetic method. Dark Fenton reaction and solar driven photo-Fenton process were used by Bandala et al. (2008) for the decolourisation of water contaminated with a benzidine-based azo dye. Using solar energy to promote the Advanced Oxidation Process (AOP) proposed, the reaction kinetic was increased twice, compared to dark Fenton reaction. With solar driven-photo Fenton, decolourisation of 90% was achieved using 1 mM of Fe^{2+} and 50 mM of H_2O_2 with only 12 kJ/L. When these conditions were tested for real wastewater, 56% of colour removal was obtained and 62.6% of the COD found initially in the effluent was eliminated after 16 kJ/L.

Hu and Ng (2008) investigated the decolorization efficiency of C.I. Reactive Red 2 (RR2) in O_3, O_3/H_2O_2, O_3/Fe^{3+}, $O_3/H_2O_2/Fe^{3+}$, UV/O_3, $UV/O_3/Fe^{3+}$, $UV/O_3/H_2O_2$ and $UV/O_3/H_2O_2/Fe^{3+}$ systems at various pHs. The experimental results indicated that the energy efficiency was highest at $[H_2O_2]_0$=1000 mg/L and $[Fe^{3+}]_0$=25 mg/L. The experimental results revealed that the combination of Fe^{3+} or H_2O_2/Fe^{3+} with O_3 at pH 4 and of H_2O_2 or H_2O_2/Fe^{3+} with UV/O_3 at pH 4 or 7 yielded a higher decolorization rate than O_3 and UV/O_3, respectively. At pH

4, the electrical energy per order of pollutant removed (EE/O) results demonstrated that the $UV/O_3/H_2O_2/Fe^{3+}$ system reduced 85% of the energy consumption compared with the UV/O_3 system.

A wastewater containing low dyestuff concentrations was treated by means of electro-Fenton process. The color was removed by in situ electrogenerated hydrogen peroxides at a three-dimensional graphite cathode with added ferrous sulfates. The removal efficiency of the color in the cathodic chamber reached 70% under specified operation conditions in 150 min. The optimal applied current density was found to be 68 A/m^2 (Wang et al., 2008). Daneshvar et al. (2008) considered the electro-Fenton treatment of Orange II producing hydrogen peroxide by oxygen reduction on graphite cathode. Experimental results showed that increasing surface area, accumulation of H_2O_2 increased and this lead to an increase in the dye decoloration. $NaClO_4$ was the best electrolyte and low concentration (0.05 M) was used. Sparging rate of 8 mL/min for 200 mL of dye solution by 7.6 cm^2 cathode surface area and dilute dye concentration were found the best conditions.

Flores et al. (2008) demonstrated that the couple Fe^{3+}/H_2O_2 was as efficient as the Fe^{2+}/H_2O_2 during a Fenton-type process. Fly ash from a coal power plant was used as iron support. Heterogeneous catalyst (Fe^{3+} - containing ashes) was stable for at least 6 h in aqueous solutions similar to those found in textile effluents. It was possible to oxidize 0.061 mM Reactive Black 5 using stoichiometric amount of H_2O_2.

Wu et al. (2008a) employed homogenous ($O_3/Mn(II)$) and heterogeneous ($UV/TiO_2/O_3$, and O_3/MnO_2) catalytic ozonation systems to decolorize C.I. Reactive Red 2 (RR2). In $O_3/Mn(II)$ system, the decolorization rate increased with concentrations of Mn(II) and MnO_2 in the ranges of 0.05-0.1 and 0.05-0.8 mg/L, respectively. In the $UV/TiO_2/O_3$, the decolorization rate increased with a TiO_2 dosage of 0.5-1 g/L and declined as the dosage increased. After 5 min reaction, the decolorization rates followed the order $O_3/Mn(II)>O_3/MnO_2>O_3>UV/TiO_2/O_3$. According to Sirés et al. (2008) aqueous single-compound and mixtures containing four thriphenylmethane (TPMs), Malachite Green (MG), Crystal Violet (CV), Methyl Green (MeG) and Fast Green FCF (FCF) were totally depolluted by electro-Fenton process with carbon-felt cathode and Pt anode. Absolute rate constants for each dye were in the $(1.31-2.65)\times10^9$ M^{-1} s^{-1} range, increasing in the order MeG<FCF<CV<MG.

Wu et al. (2008b) employed Mn(II), Fe(II), Zn(II), Co(II) and Ni(II) ions to catalyse ozonation and increase its decolorization ability versus Reactive Red 2 (RR2). They observed that, in metal-catalyzed ozonation, decolorization was better at pH 2 than at pH 5, except in the $O_3/Fe(II)$ system. Under all the experimental conditions tested, the decolorization efficiency of catalytic ozonation exceeded that of ozonation alone. In their paper, Martínez et al. (2008) refer that the Cu(II)-poly(EDGE-MAA-2MI) complex acid acted as an efficient heterogeneous catalyst in the degradation of H_2O_2 with decolorization of the azo-based dye Methyl Orange (MO). The catalyst showed to be

stable at initial hydrogen peroxide concentrations up to 50 mM. Besides, its efficiency increased with the amount of immobilized Cu(II). Combining an adequate ratio of mass of complex to solution volume with 25 mM $[H_2O_2]_0$, an almost complete removal of color was achieved between 10 and 20 min, in a neutral medium at room temperature.

Decolorization and mineralization of the non-biodegradable azo dye Orange II (OII) solutions by solar photo-Fenton (SPF) and ferrioxalate-assisted solar photo-Fenton (SPFox) reactions were carried out by Monteagudo et al. (2008) in a solar compound parabolic collector (CPC). The results revealed that the addition of oxalic acid to the Fenton system ($Fe(II)/H_2O_2$) irradiated under sunlight improved the photocatalytic efficiency. Under the optimal conditions, 100% decolorization of dye solution could be reached by using both processes, but with different accumulated solar energy. However, the efficiency of TOC and Chemical Oxygen Demand (COD) removal was higher in the SPFox. Daneshvar et al. (2008) found that UV/H_2O_2 process was a powerful method for decolorization of Rhodamine B (RB). For 10 mg/L RB concentration, an optimum dose of H_2O_2 (450 mg/L) at the light intensity of 29.7 W/m^2 was found to give a rate constant of 0.0409 min^{-1} and 0.085 min^{-1} for decolorization and degradation, respectively.

Electrochemical. Körbathi and Tanyolaç (2008) studied the electrochemical oxidation of simulated textile wastewater on iron electrodes in the presence of NaCl electrolyte in a batch reactor. The highest COD, color and turbidity removals were achieved as 93.9%, 99.5% and 82.9%, respectively, at 40% pollution load, 8 V applied

potential, 37.5 g/L electrolyte concentration and 30°C reaction temperature. The treatment was optimized by using the RSM.

Degradation of cationic red X-GRL by electrochemical oxidation on a novel PbO_2 anode modified by fluorine resin was investigated by Zhou and He (2008). They observed that current density, temperature and initial dye concentration significantly affected color and COD removal. Current density of 2.0 mA/cm^2 was chosen to be the best value. The process showed a high current efficiency and competitive energy consumption for effective treatment of dye wastewater containing a certain salt content.

Electrochemical treatment was applied to the treatment of a synthetic solution containing Acid Blue 22 using a boron-doped diamond electrode (BDD). During oxidation, development of a polymeric film in the potential region of water stability was observed which caused electrode fouling; it was removed by high-potential anodic polarization in the region of O_2 evolution. Complete COD and color removal were achieved within the investigated range of conditions (Panizza and Cerisola, 2008). A pilot-scale electrochemical reactor was set-up by Agarwal et al. (2008) to study degradation of calmagite, a sulfonated azo dye used as a model contaminant. Treatment involved recirculating the contaminated solution through the electrode beds at small flow velocities. The results showed hydraulic stability, consistent pH behavior, marginal temperature rise (<5°C) and overall safe and predictable performance under diverse conditions. Near complete removal of calmagite was seen at 3-10 V of applied voltage

in 8-10 h. Wu et al. (2008) used a three-dimensional electrode reactor with granular carbon aerogels (Cas) as particle electrodes to experimentally investigate decoloration of RBRX from simulated dye wastewater. The results showed that decoloration ratio was still as high as 95% after more than 100 h treatment run. The decolorizing rate increased with the increase of treating time, cell voltage and particle electrode dose, but decreased with initial dye concentration.

Decolourisation of real textile effluent by constant current electrolysis in a flow-cell using DSA® type material was studied by Malpass et al. (2008). Total removal of color was possible when NaCl was added to the electrolyte and high current densities were employed. When the *as-received* effluent was treated without the addition of NaCl, both color and TOC removal were limited. The best results in this case were obtained when $Ti/Ru_{0.1}Sn_{0.9}O_2$ electrode was used.

Coagulation-Flocculation. The use of dissolved chitosan for the removal of sulphonated azo dyes (Acid Black 1, Reactive Black 5, Acid Violet 5) from acidic solutions was studied by Szygula et al. (2008). The number of sulphonic groups on the dye was correlated to the effectiveness of the coagulation-flocculation process, and it was found that the molar ratio between the dye molecules and the amine groups respected the stoichiometry between sulphonic functions and protonated amine groups only in the case of Acid Violet 5. Optimal pH and coagulant dosages were determined. Aleboyeh et al. (2008) investigated the decolorization of C.I. Acid Red 14 (AR14) azo dye by electrocoagulation (EC) process in a batch reactor. The optimum operational conditions were found through RSM to be: 102 A/m^2, 4.47 min and 7.27, respectively. Under these conditions, AR14 removal higher than 91% was attained. Zidane et al. (2008) showed the possibility to use mineral coagulants, previously produced by electrolysis of saline solutions, using aluminium electrodes to remove C.I. Reactive Red 141 dye pollutant effluent. The best performance of dye removal were obtained with the inorganic coagulant C_2 produced by means of electrolysis of sodium chloride solution, S_2 [NaCl $(10^{-2}$ M)] resulting in a mixture of polymeric species $(Al_{45}O_{45}(OH)_{45}Cl)$ and monomeric species $(AlCl(OH)_2 \cdot 2H_2O$ and $Al(OH)_3)$. The removal efficiency evaluated by measuring the yields of 540-nm absorbance varied from 41% to 96% through 60 min of treatment by using C_2 concentrations ranging from 100 to 400 mg/L.

Cañizares et al. (2008) proposed a model for the electrocoagulation process and then applied it to simulate the treatment of wastes polluted with Eriochrome Black T (EBT) solutions. For the treatment of EBT solutions, the enmeshment of EBT in a growing aluminium hydroxide precipitate and the charge neutralization of EBT molecules were considered as primary mechanisms. Yildiz (2008) applied Taguchi method to determine the optimum working conditions for the dye removal (Bomaplex Red CR-L) from aqueous solutions by electrocoagulation. The optimum conditions were found to be: initial dye concentration of 100 mg/L, initial pH of the solution of 3, supporting electrolyte concentration of 0.0 mM, supporting electrolyte type of $CaCl_2$ and current density of 0.050 mA/cm^2.

Polyamine flocculants were synthesized by the polycondensation of dimethylamine and epichlorohydrin, in which organic amines were used as modifying agents. Their flocculation performance was evaluated with simulated dye liquor and actual printing and dyeing wastewater. The highest efficiency of color and COD removal from polyamine for treating dye wastewater was 90% and 89%, respectively.

Membrane. Criscuoli et al. (2008) demonstrated feasibility of vacuum membrane distillation (VMD) for concentrating solutions containing different amount of dyes (25-500 mg/L) while recovering pure water at the distillate side. The permeate flux increased with feed temperature and flow rate. Decay of permeate flux due to fouling was registered only in the first 30 min of run, after which an asymptotic value was reached.

Djenouhat et al. (2008) studied the utilization of water-in-oil (W/O) emulsions prepared by low-frequency ultrasonic irradiation (22.5 kHz) for the removal and recovery of cationic dyes from aqueous solution by emulsion liquid membrane containing di(2-ethylhexyl) phosphoric acid (D2EHPA) as carrier. The results obtained showed that, under most favourable conditions, practically all the cationic dyes present in the feed phase were extracted. The removal percentage of the mixed dye was found to be higher than 98% even in the presence of salt in the continuous phase. The best sulphuric acid concentration in the internal phase that conducted to very good stripping efficiency (above 97%) and excellent emulsion stability was 0.5 M.

Extraction of dyes from aqueous solutions was also studied by Das et al. (2008) using liquid emulsion membrane, made by *n*-heptanes as membrane phase, sodium hydroxide solution as internal phase and dye solution as external phase. Maximum extraction for Methylene Blue was found to be 99% and for Crystal Violet about 95% in single component system. In binary mixture these values were 97% and 90%, respectively.

A biologically treated wastewater from a cotton thread factory was subjected to nanofiltration (NF) in two ways, direct NF treatment and NF after a pre-treatment by ultrafiltration (UF). Three different NF membranes of different pore size (NF90, NF200 and NF270 from Dow-Filmtec) were studied. Experiments were carried out at four different pressures in a pilot plant equipped with a flat-sheet membrane module. The NF90 yielded a COD reduction of 90% and the highest salt retention (75-95%), which allowed to meet specifications for water reuse in the textile industry. Pre-treatment with UF increased permeate flux of NF of about 50% and reduced COD concentration in NF feed of about 40% (Gonzálvez-Zafrilla et al., 2008). A comparison of direct NF and combined UF/NF was also carried out by Fersi and Dhahbi (2008). Experimental results showed that the combination of UF with NF improved the textile permeate quality by increasing the retention values of the majority of analyzed parameters. For instance, color retention of about 95%, conductivity and total dissolved salts retentions of about 80% and bivalent ions retention above 95% were achieved. According to Avlonitis et al. (2008), TRISEP (4040-XN45-TSF) NF membranes showed excellent decolorization efficiency of

cotton dye effluents, combined with adequate salt rejection properties (more than 72%) at reasonable energy consumption. Capar et al. (2008) studied the recovery of print- and beck-dyeing wastewaters of the carpet manufacturing industry by membrane processes. A combined treatment after separate pre-treatment stages was adopted to simplify the overall process. The results showed that these wastewaters can be treated together to the degree of reuse quality in a single NF unit even when mixed at equal volumes.

Adsorption. Gerçel et al. (2008) demonstrated the ability of using activated carbon prepared from *Euphorbia rigida* by sulfuric acid chemical activation for the adsorption of Disperse Orange 25 (DO25) from aqueous solutions. The amount of dye adsorbed was found to be dependent on solution pH, adsorbent concentration, initial dye concentration, contact time and temperature. The highest adsorption capacity was found to be 118.93 mg/g at 20°C and pH around 7. Equilibrium data fitted well in a Langmuir isotherm equation.

The biosorption potential of a fermentation industry waste (*Corynebacterium glutamicum*) towards Methylene Blue (MB) was enhanced via succination, which chemically modified the amine groups to carboxyl. Both raw and succinated *C. glutamicum* performed well in MB biosorption at neutral or basic conditions. The succinated biomass outperformed raw biomass in MB biosorption but underperformed compared to two commercial activated carbons. Kinetic experiments revealed that almost complete biosorption equilibrium was attained within 5 min,

followed by complete saturation in 30 min, for both forms of *C. glutamicum* (Vijayaraghavan et al., 2008).

A series of surfactant-modified montmorillonites (MMT) were prepared by Wang and Wang (2008) and used for the adsorption of Congo Red (CR) anionic dye from aqueous solution. Compared to MMT, the adsorption capacity of surfactant-modified montmorillonites MMT for CR was greatly enhanced and MMT modified with cetyltrimethylammonium bromide (CTAB) exhibited the higher adsorption capacity. According to Jin et al. (2008), 2% sodium dodecyl benzenesulfonate (SDBS)- and 3% sodium dodecyl sulfate (SDS)-modified zeolites had higher adsorption capacities for Methylene Blue than the unmodified zeolite, while 2% cetylpyridinium bromide hexadecyl (CPB)- and 2% hexadecylammonium bromide (HDTMA)-modified zeolites were the best adsorbents for Orange II.

A new isotherm equation, the Brouers-Sotolongo isotherm (BSI), a deformed (Weibull) exponential equation was used by Ncibi et al. (2008) to describe adsorption equilibrium of phenol and Methylene Blue (MB), respectively onto a non-porous adsorbent, *Posidonia oceanica* fibres and two porous adsorbents, chemically and physically activated carbons (ACs) prepared from vetiver roots. The adsorption characteristic parameters obtained were also calculated using classical adsorption isotherms, i.e. Langmuir isotherm (LI), Freundlich isotherm (FI) and Redlich-Peterson isotherm (RPI). For *Posidonia* and as well for the ACs, when MB was the adsorbate, the RPI was the best-fitting model. Roulia and Vassiliadis (2008) investigated the adsorption of C.I. Basic Blue 41 onto raw

perlite, expanded perlite, bentonite and montmorillonite. The clays showed to be more efficient than perlites but their dye retention was less increased with pH and temperature. The Langmuir and Harkins-Jura isotherms were found to better model experimental data from clays while the Freundlich, Halsey, Henderson, Brunauer-Emmett-Teller and Smith models fitted more successfully data from the application of perlites.

Various binding mechanisms for the uptake of reactive dyes (Reactive Blue 4, RB4, Reactive Orange 16, RO16, Reactive Yellow 2, RY2) by the protonated waste biomass of *Corynebacterium glutamicum* were investigated by Won et al. (2008). At acidic pH, the electrostatic interaction was found to be a major binding; under alkaline conditions, the binding mechanisms were quite different according to the reactivity of dyes. Engin et al. (2008) applied Taguchi method to determine optimum conditions for color removal from textile dyebath house effluents in a zeolite fixed bed reactor. In order to increase the adsorption capacity, the surface of natural zeolite was modified with a typical quaternary amine surfactant, hexadecyltrimethylammonium bromide (HTAB, $C_{19}H_{42}BrN$). The optimum experimental conditions were found to be as follows: HTAB concentration =1 g/L, HTAB feeding flowrate=0.015 L/min, textile wastewater flow rate =0.025 L/min and bed height=50 cm. Under these conditions, the treated wastewater volume reached a maximum while the bed volumes were about 217.

Unloaded polyether type polyurethane foam was used by Baldez et al. (2008) as an effective adsorbent for the removal of Methylene Blue (MB) from waters in a medium containing dodecylsulfate anion (SDS). Better results were obtained when SDS molar concentration was five times higher than MB. The amount of dye adsorbed increased with the increase of temperature, indicating that the adsorption was an endothermic process.

Granules prepared from dried activated sludge (DAS) were utilized as a sorbent for the uptake of Rhodamine-B (Rh-B) dye. Nearly 15 min was required for the equilibrium adsorption, and Rh-B dyes could be removed effectively. The acid pretreated biomass exhibited a slightly better biosorption capacity than alkali pretreated or non-pretreated biomass (Ju et al., 2008). Adsorption studies for the removal of C.I. Acid Blue (AB25) from aqueous solutions onto a cationized starch-base material were performed by Renault et al. (2008). The process was uniform and rapid and the adsorption kinetics followed the pseudo-second order model. The Langmuir equation provided an accurate description of the experimental data.

Tan et al. (2008) studied the effect of hydrochloric acid (HCl) treatment of activated carbon prepared from oil palm shell on Methylene Blue (MB) adsorption. The equilibrium data for both the untreated and HCl-treated activated carbons were best represented by the Langmuir isotherm. The adsorption capacity of HCl-treated activated carbon was 303.03 mg/G, which was 24.24% higher compared to that of the untreated activated carbon. According to Yener et al. (2008), sorption capacity of Methylene Blue on powdered activated carbons (PAC) was much higher than on granular activated carbon (GAC). In the case of using GAC with particle size of 0.0864 cm, pore diffusion was found to be the controlling mechanism for

adsorption rate. The adsorption of Malachite Green (MG) on sodium-exchanged kaolinite and on the same clay treated with two widely used deflocculants, sodium hexametaphosphate (NaHMP) and sodium disilicate (NaDIS), was investigated by Castellini et al. (2008). The results obtained indicated that the deflocculants affected the adsorption process. In particular, DIS treatment increased physisorption, whereas HMP treatment almost completely prevented it.

The capability of three selected fungi, *Rhizopus arrhizus, Trametes versicolor, Aspergillus niger*, for removing Gryfalan Black RL metal-complex dye was examined by Aksu and Karabayir (2008). The maximum specific sorption capacity determined according to the Langmuir model was as follows: *R. arrhizus* (666.7 mg/g at pH 2.0 and 25°C)> *A. niger* (500.0 mg/g at pH 1.0 and 35°C)>*T. versicolor* (434.7 mg/g at pH 2.0 and 25°C). The rate of biosorption was very rapid for each adsorbent attaining equilibrium within 4-6 h. Continuous mode sorption experiments were carried out by Rastogi et al. (2008) to remove Methylene Blue from aqueous solutions in hydrocyclone equipment onto fly ash. A maximum removal of 58.2% was observed at adsorbent dosage of 900 mg/L at pH 6.75 for an initial MB concentration of 65 mg/L. Hameed et al. (2008) determined sorption isotherm of MB onto banana stalk waste at 30°C with initial concentration of MB in the range of 50-500 mg/L. At pH 2.0, the sorption of dye was not favorable, while the sorption at other pHs (4.0-12.0) was remarkable. Equilibrium data were best represented by the Langmuir

isotherm model, with maximum monolayer adsorption capacity of 243.90 mg/g.

Adsorption of Malachite Green (MG) onto bentonite in a batch adsorber was seen to increase with increasing contact time until equilibrium and initial dye concentration. The adsorption capacity of bentonite was independent on initial pH in the range of 3-11 (Bulut et al., 2008a). The reductive decolorization of the textile dye Reactive Blue 4 (RB4) using zero-valent iron (ZVI) filings was evaluated by Epolito et al. (2008) in batch assays. The decolorization rate increased with decreasing pH and increasing temperature, mixing intensity, and addition of salt (100 g/L NaCl) and base (3 g/L Na_2CO_3 and 1 g/L NaOH) conditions typical of textile reactive dyebaths.

In their experiments, Lin et al. (2008) observed that degradation efficiency of AB24 dye in aqueous solution with nano/micro-size zero valent iron (ZVI) increased with increasing ZVI concentration and temperature but decreased with particle size of ZVI. The activation energy of reaction was 72.3 kJ/mol, indicating that surface reaction controlled the rate of degradation. Three organic dyes, namely Coomassie Blue (CB), Malachite Green (MG) and Safranin Orange (SO), were removed from solution by adsorption on sand at 298K. Under optimized conditions, 65-70% of dye could be removed from solution onto the sand surface. Adsorption data was fitted to Freundlich equation while adsorption kinetics followed the pseudo-second order equation for all the three dyes investigated with the k value lying in the region of 6.2×10^5 to 3.0×10^6. Malachite Green reached equilibrium faster than the other dyes (Rauf et al., 2008).

The effect of temperature on the adsorption of Metamil Yellow (MY) (acidic) and Methylene Blue (MB) (basic) by poplar sawdust was investigated by Pekkuz et al. (2008). In addition, amounts of $NaHCO_3$, Na_2CO_3, NaOH and C_2H_5ONa adsorbed by 1 g of poplar sawdust were determined. Results showed that poplar sawdust adsorbed cationic dyestuffs more than anionic dyestuffs. Calcified eggshell (ES) and its ground eggshell powder (ESP) were used by Tsai et al (2008) for the removal of cationic Basic Blue 9 (BB9) and anionic Acid Orange 52 (AO52) from aqueous solution. The adsorption potential for BB9 onto ESP was far lower than that for AO52, mainly due to the anionic interaction between the acid dye with the sulfonate groups and the positively charged sites on the surface of ESP. Besides, the adsorption capacity of AO52 onto the calcified eggshell was significantly smaller that that onto its ground form. An increase in adsorption temperature from 15 to 45°C significantly enhanced the adsorption capacity of AO52 onto ESP.

Uğurlu et al. (2008) compared the adsorptive capacity of commercial activated carbon and activated carbon prepared from olive stone for the removal of Remazol Red B (RRB) from aqueous solutions. Different activating agent ($ZnCl_2$) amounts and adsorbent particle size were studied to optimise adsorbent surface area. According to the results obtained, the equilibrium time, optimum pH, adsorbent dosage were found: 60 min, pH<3-4 and 1.0 g/50 mL, respectively. Lower adsorption capacity for RRB on activated carbon prepared from olive stone was observed. Hsu (2008) found that raw coal fly ash (CFA) that had not been subjected to any pre-treatment process,

had superior adsorbing ability for the anionic dye Acid Red 1 (AR1) dye than did two modified coal fly ashes (CFA-600 and CFA-NaOH). The adsorption capacity followed the order CFA>CFA-600>CFA-NaOH and they each increased upon increasing the temperature.

The adsorption kinetics of a cationic dye, Methylene Blue (MB), onto the silica nano-sheets derived from vermiculite via acid leaching was investigated by Zhao et al. (2008). Experimental results showed that increasing initial dye concentration favored the adsorption while the acidic pH and temperature influenced negatively. Data were well fitted by the pseudo-second order kinetic model. The activation energy was calculated as 3.42 kJ/mol.

Bottom Ash and deoiled soya were effective in removing Brilliant Green from water. For both adsorbents, the adsorption process was found to be dependent on pH of the dye solution, concentration of the dye, amount of sieve sizes of the adsorbents, temperature. For deoiled soya a film diffusion mechanism operated at all the temperatures. (Mittal et al., 2008). Hameed et al. (2008) demonstrated the ability of coconut bunch waste (CBW) to remove basic dye Methylene Blue (MB) from aqueous solution by adsorption. Equilibrium isotherms were well described by the Langmuir equation, giving maximum adsorption capacity of 70.92 mg/g at 30°C. The use of a low-cost and eco-friendly adsorbents made by activated carbon prepared from agricultural waste materials such as gingelly (sesame) (Sp), cotton (Cp) and pongam (Pp) seed shells was investigated by Thinakaran et al. (2008) for the removal of Acid Red 114 (AR114) from aqueous solutions. Optimum

conditions were found to be pH 13, adsorbent dosage of 3 g/L of solution and equilibrium time f 4 h. Higher removal percentages were observed at lower concentrations of AR114. Better performances were obtained with Pp seed shells as compared to Cp and Sp seed shells.

According to Gong et al. (2008), citric acid esterifying wheat straw (EWS) was an excellent adsorbent for removal of cationic dyes, MB and Crystal Violet (CV). The optimal pH value and EWS dosage for favorable adsorption were found to be pH 14 and above, and 2 g/L for 250 mg/L of dye solution, respectively. At this dosage, the adsorption percentages of MB and CV kept above 95% over a range from 50 to 350 mg/L of dye concentration.

Köse (2008) prepared an anion exchanger resin from agricultural residue (straw) after reaction with epichlorohydrin and dimethylamine in the presence of pyridine and *N,N*-dimethylformamide (EDM) method. The new adsorbent material was used for removal of Remazol Black B dyestuff from wastewater in a batch system. The applied statistical analysis showed that sorption was favoured by increase in initial concentration and temperature and unfavoured by and increase in initial pH of wastewater.

Novel *N,O*-carboxymethyl-chitosan/montmorillonite (*N,O*-CMC-MMT) nanocomposites were synthesized by intercalation reaction between *N,O*-carboxymethyl-chitosan (CMC) and montmorillonite (MMT) in distilled water. Adsorption tests of Congo Red (CR) on *N,O*-CMC-MMT nanocomposites were carried out and the results obtained from the study showed that the nanocomposite whit the molar ratios of

N,O-CMC to MMT of 5:1 exhibited higher adsorption capacity (Wang and Wang, 2008).

In their study, Hamdaoui et al. (2008) demonstrated that dead leaves of plane tree can be successfully used for the removal of hazardous dye, Malachite Green (MG), from aqueous solutions. The batch sorption process was found to be dependent upon contact time, sorbent dose, initial dye concentration, ionic strength and temperature. The obtained value of the activation energy was very low (7.13 kJ/mol), which indicated not only an activated process but a physical sorption.

Ofomaja (2008) studied the adsorption of Methylene Blue (MB) and Methyl Violet (MV) using mansonia wood sawdust as adsorbent. Maximum dye removal was observed at pH 10 for both dyes. The pseudo-second order kinetic was found to fit the experimental data and was able to consistently predict the amount of dye adsorbed over the sorption period and for the range of sawdust particle size used. Andini et al. (2008) demonstrated that MB can be effectively adsorbed onto fuel oil fly ash, without any pre-treatment. The adsorption capacity was found similar to that of other carbon-rich waste-based adsorbents. Besides, fuel oil fly ash showed to exert high buffering capacity. Unuabonah et al. (2008) used raw kaolinite clay and its sodium tetraborate (NTB)-modified analogue to adsorb Aniline Blue dye. The modification increased the adsorption capacity of the adsorbent from 1666.67 to 2000 mg/kg for Aniline Blue dye, and also its initial adsorption rate for the dye. Increase in pH and adsorbent weight increased the dye adsorption onto the unmodified sorbent. However, for the NTB-

modified sorbent, increasing pH did not improve the adsorption capacity but increased its overall pseudo-second order rate constant. Activated carbons were prepared by Hameed and El-Khaiary (2008) from bamboo by chemical activation with K_2CO_3 and physical activation with CO_2 (BAC) for the removal of Malachite Green (MG). The Langmuir isotherm was found to have the best fit to the experimental data, with maximum adsorption capacity of 263.58 mg/g.

Khalaf (2008) demonstrated the ability of the inactive biomass of *Aspergillus niger* and *Spirogyra* sp. to remove reactive dye (Synazol) from multi-component textile wastewater. Pre-treatment of fungal and algal biomasses with autoclaving increased the dye removal more than pre-treatment with gamma-irradiation. Dried autoclaved biomass of *A. niger* and *Spirogyra* sp. exhibited maximum dye removal (88% and 85%, respectively) at pH 13, temperature 30°C and 8 g/L (w/w) biomass concentration after 18 h contact time. A practical biosorbent for the decolorization of textile effluents was developed by Vijayaraghavan et al. (2008) from the fermentation waste, *Corynebacterium glutamicum* biomass. This product, when decarboxylated and immobilized in polysulfone matrix, performed well in decolorization of four different reactive dyes and other auxiliary chemicals. The regeneration of polysulfone-immobilized *C. glutamicum* was successful with the aid of 0.01 M NaOH as the eluant, which enabled the biosorbent to maintain consistent decolorization efficiency for up to 25 cycles. Activated carbons were produced from sunflower oil cake by sulphuric acid activation with different impregnation

ratios by Karagöz et al. (2008). The experimental impregnation ratio strongly affected the adsorption capacity and the optimum value was found to be 0.85. The effects of various process parameters were investigated. The optimum conditions were found to be 25°C and pH 6 for the maximum adsorption of MB. Ofomaja and Ho (2008) demonstrated that biosorption of Methyl Violet (MV) from aqueous solution by Mansonia wood sawdust is a spontaneous and endothermic process. The saturated monolayer biosorption capacity of the Mansonia wood sawdust for MV at 329K was calculated to be 24.6 mg/g. Desorption of biosorbed MV dye increased with increasing HCl concentration, suggesting that the removal mechanism was by ion exchange.

Apricot stone with H_2SO_4 activation was used as adsorbent to remove a basic dye, Astrazon Yellow 7GL from aqueous solutions. The optimum conditions for removal were found to be pH 10, 6 g/L adsorbent dosage and equilibrium time of 35 min. The removal rate was heavily dependent on diffusion controlled kinetic model. The adsorption capacity was calculated as 221.23 mg/g at 50°C (Demirbas et al., 2008). A series of novel sodium humate/poly(*N*-isopropylacrylamide) (SH/PNIPA) hydrogels were synthesized by solute polymerisation. The adsorption and desorption of MB for the hydrogels were influenced by temperature, initial MB concentration and SH amount. Low temperature favored the adsorption and desorption of MB. The maximum adsorption capacity was 10.8 mg/g (Yi et al., 2008).

Wu and Tseng (2008) investigated the adsorption equilibrium of Acid Blue 74 (AB74), Basic Blue 1 (BB1)

and Methylene Blue (MB) on NaOH-activated carbons (FWNa2, FWNa3, and FWNa4) through the surface coverage ratio (S_c/S_p) and monolayer cover adsorption amount per unit surface area (q_{mon}/S_p). The q_{mon}/S_p of NaOH-activated carbons was better than that of KOH-activated carbons prepared from the same raw material (fir wood). The S_c/S_p values of the adsorption of all adsorbates on adsorbent FWNa3 were found to be higher than those in related literature. Hameed and Hakimi (2008) evaluated ability of durian peel (DP), an agricultural waste, to remove Acid Green 25 (AG25) from aqueous solutions. They found that sorption equilibrium was best described by the Langmuir isotherm model with maximum monolayer adsorption capacity of 63.29 mg/g at 30°C. Kinetic studies indicated that the sorption of AG25 tended to follow pseudo-second order kinetic.

Adsorbents prepared from parthenium weed by the chemical treatment, i.e. phosphoric acid treated parthenium carbon (PWC) and formaldehyde treated parthenium biomass (WC), were used by Lata et al. (2008) for the removal of Rhodamine-B (RB) over a wide range of concentrations. The results indicated that WC had lower adsorption efficiency (63.16%) for RB than PWC (85.95%) under studied experimental conditions. Higher dye adsorption was possible provided initial dye concentration was low in solution According to Du et al. (2008), chitosan nanoparticles showed an excellent capacity for adsorption of anionic dye, Eosin Y, due to the large relative surface area and the high zeta potential. The adsorption process was very fast and attained equilibrium within 4 h. The adsorbed dye could be desorbed efficiently using alkaline solution.

Chitosan-based adsorbent exhibited interesting sorption properties toward Basic Blue 3 (BB3) (the maximum adsorption capacity was 166.5 mg/g), depending on the presence of sulfonate groups (Crini et al., 2008).

Solid wastes (SW) from distillery, a by-product of the ammonia-soda process (the Solvay method) for the production of soda ash, were used by Şener (2008) for removing the anionic reactive dye Procion Crimson H-EXL (Reactive Red 231, RR231). The results indicated that the adsorption was strongly dependent on pH. Particularly, as the pH increased, extent of dye uptake increased and $Ca(OH)_2$ particles precipitated at higher pH were mainly responsible for the removal of the dye. The necessary time to reach equilibrium was found to be less than 2 min.

The crude dehydrated sewage sludge from an urban wastewater treatment plant showed to have the capacity to fix 19.6 mg/g Direct Red 79 and 248.3 mg/g Vat Blue 4 in synthetic effluents. When used to treat a real textile mill effluent, the adsorbent showed that 8 mg/g of the dominant dye (Indigo) was adsorbed (Dhaouadi and M'Henni, 2008). Dried biomass of *Caulerpa racemosa* var. *cylindracea*, an invasive species in the Mediterranean Sea, was successfully used by Cengiz and Cavas (2008) in the removal of Methylene Blue. The adsorption reached equilibrium at 90 min for all studied concentrations (5-100 mg/L). There was a sharp increase in the adsorbed dye amount per adsorbent amount from 3.3 to 16.7 g/L, then a slight increase up to 66.7 g/L was observed. The yellow passion fruit (*Passiflora edulis* Sims. f. *flavicarpa* Degener)

peel, a powdered solid waste, was tested by Pavan et al. (2008) as an alternative low-cost adsorbent for the removal of a basic dye, Methylene Blue (MB). The contact time required to obtain the maximum adsorption (0.0068 mmol/g) was found to be 56 h at 25°C and pH 9.0.

Akar et al. (2008) refer that biosorption of Acid Blue 40 (AB40) onto cone biomass of *Thuja orientalis* was fast and the equilibrium was attained within 50 min. Maximum biosorption capacity for AB40 was 97.06 mol/g at 20°C. Biosorption of Congo Red (CR) onto rice husk in a continuous fixed bed was studied by Han et al. (2008). Data confirmed that breakthrough curves were dependent on flow rate, initial dye concentration and bed depth. The initial region of the breakthrough curves was defined by the Bohart-Adams model at all the experimental conditions studied while the full description of breakthrough could be accomplished by Thomas models. Cheng et al. (2008) used native and heat-treated anaerobic granular sludge in removing Malachite Green (MG) from aqueous solutions. The maximum adsorption was observed at pH 5.0 on both native and heat-treated sludge. Kinetic studies showed that the biosorption process followed pseudo-second order and the maximum adsorption on native and heat-treated sludge was found to be 61.73 and 59.17 mg/g at initial concentration of MG of 150 mg/L, respectively.

Activated carbons were prepared from rubber (*Hevea brasiliensis*) seed coat and used to remove Basic Blue 3 (BB3) from aqueous solutions. The equilibrium adsorption was best represented by the Freundlich isotherm. The monolayer adsorption capacity was 227.27 mg/g at 30°C (Hameed and Daud, 2008). The adsorption of

Acid Red 14 (AR14) and Acid Blue 92 (AB92) onto the microporous and mesoporous egg shell membrane (ESM) was investigated by Aramai et al. (2008) in aqueous solution in a batch system with respect to different parameters. The surface area of ESM was found to be 2.2098 m^2/g. The biosorbent exhibited high sorption capacity for both dyes Desorption studies were also conducted.

Crab shell powder was able to effectively remove the colour from textile wastewater. The application of direct solid-phase microextraction (SPME) showed to be a suitable methodology for the determination of the nine compounds in the textile effluent (Sye et al., 2008). The study of Pengthamkeerati et al. (2008) investigated the adsorption behaviour of Reactive Black 5 (RB5) and Reactive Yellow 176 (RY176) from aqueous solution on coal fly ash (FA-CO), HCl-treated coal fly ash (TFA-HCL), and biomass fly ash (FA-BM). Preliminary studies showed that FA-BM had the greatest dye adsorption capacity of both dyes. Hence, only for the FA-BM, the effects of various experimental parameters were determined. At the final pH of 8.1-8.5, the adsorption capacity of both dyes on FA-BM was maximum and decreased above or below this pH. A positive effect of salt addition on the dye adsorption capacity was also observed. An industrial waste sludge mainly composed by metal hydroxides was used by Santos et al. (2008) for removing a reactive textile dyes (Remazol Brilliant Blue) in solution. Dye adsorption equilibrium isotherms were determined at 25 and 35°C and pH of 4, 7 and 10, revealing reasonably fitting to Langmuir and

Freundlich models. At 25°C and pH 7, Langmuir indicated a maximum adsorption capacity of 91.0 mg/g.

Dodecylsulfate- and dodecyl benzene sulfonate-hydrotalcites were prepared by calcination-reydratation method. The resulting material was characterized and used to remove a basic dye (Saframine) from aqueous solutions. The kinetic and sorption data well fitted the second order kinetic model and the Langmuir model, respectively (Bouraada et al., 2008). Prigione et al. (2008) evaluated dye decolourisation, COD and toxicity decrease of three wastewater models after the treatment with inactivated biomasses of three Mucorales fungi cultured on two different media. Fungal biomasses displayed good sorption capabilities giving rise to decolourisation percentages up to 94% and decrease of COD up to 58%.

The study of Alkan et al. (2008) demonstrated that sepiolite can be used as an adsorbent for the removal of Maxilon Blue 5G from aqueous solution. The adsorption process equilibrium attained within 60 min. The extent of dye removal increased with increasing initial concentration of dye, contact time, pH and temperature, and decreased with increasing ionic strength. The diffusion coefficient was calculated and found to be in the range of 3.625×10^{-8} to 12.100×10^{-9} cm^2/s. Batch experiments were conducted by Sun et al. (2008) to study the biosorption of a cationic dye, Malachite Green (MG), onto aerobic granules. The results showed that alkaline pH was favourable for the biosorption of MG and chemisorption seemed to play a major role in the removal process. The equilibrium time was 60 min for both 50 and 60 mg/L and 120 min for both 70 and 80 mg/L MG concentrations, respectively.

Removal of Methylene Blue (MB) was carried out using chitosan-*g*-poly(acrylic acid)/montmorillonite nanocomposites. The adsorption behaviour of the nanocomposites showed that the adsorption kinetics and isotherms were in good agreement with pseudo-second order equation and the Langmuir equation, respectively (Wang et al., 2008). Bulut et al. (2008b) conducted kinetic and equilibrium studies on the adsorption of Congo Red (CR) onto bentonite in the concentration range of 75-300 mg/L at pH 6.8 and 298K. The Langmuir isotherm was demonstrated to provide the best correlation for the adsorption of CR onto bentonite. The adsorbed amounts of CR increased with increase in contact time and attained the equilibrium after 60 min.

The beer brewery waste was shown to be a low-cost adsorbent for the removal of basic dye as compared to its precursor (i.e. diatomite) based on its physical and chemical characterization. The pore properties of this waste were significantly larger than those of its raw material, reflecting that the trapped organic matrices contained in the waste probably provided additional adsorption sites and/or adsorption area (Tsai et al., 2008).

The surface chemistry of maize natural adsorbent was altered by the modifying agents phosphoric acid and different amines (trethanolamine, diethylenetriamine and 1,4-diaminobutane). Removal of Methyl Orange (MO) (25 mg/L) was below 50% by maize corn cobs modified by phosphorylation and higher by the quaternized samples: 68% with the 1,4-diaminobutane and 73% with the diethylenetriamine modificators (Elizalde-González et al., 2008).

Coffe husks (CH), a coffee processing residue, were used by Oliveira et al. (2008) as untreated sorbents for the removal of Methylene Blue (MB). The effect of solution temperature, pH, sorbent dosage and contact time on MB removal were investigated. The experimental adsorption equilibrium data fitted to both Langmuir and Freundlich adsorption models. Oladoja et al. (2008) investigated the potentialities of castor seed shell (CSS), a waste agricultural by-product, in the remediation of water contaminated with Methylene Blue (MB). The CSS was ground and washed, thoroughly, to remove any water extractable constituents. The dried CSS was reground, sieved and used in series of agitated batch experiments. The sorption capacity of the CSS, as obtained from the Langmuir plot, was 158 mg/g. Aksu et al. (2008) used dried *Trametes versicolor*, a white-rot fungus and chitosan, a fungal component derivative, for the removal of Acid Blue 161 (AB161) and compared the results obtained with the outcomes of acid-washed powdered activated carbon (PAC). All sorbents exhibited the highest dye uptake capacity at initial pH value of 3.0. The effect of temperature indicated that maximum capacity was obtained at 45°C for each AB161 dye-sorbent system. Sorption capacity of each sorbent increased with increasing initial dye concentration up to 500 mg/L. Among the three adsorbents, chitosan was the most effective showing a maximum dye uptake of 471.6 mg/g at 45°C. The equilibrium adsorption of C.I. Acid Blue 25 by a cationized starch-based (MRS) adsorbent was reported by Gimbert et al. (2008). This material exhibiting an interesting capacity although the value was temperature-dependent. According

to Gong et al. (2008), rice straw esterified thermochemically with citric acid (CA) was an excellent Methylene Blue sorbent. The optimal pH was 3 and above. For the 250 mg/L MB solution, 1.5 g/L or up of the sorbent could almost completely remove the dye from aqueous solutions.

Dead *Penicillium chrysogenum* mycelium was used for the removal of three anionic dyes: Acid Orange 8 (AO8), Acid Blue 45 (AB45) and Reactive Orange 16 (RO16). The biomass surface was modified with polyethylenimine (PEI) and cross-linked with glutaraldehyde for enhanced anionic dye sorption. Compared to the pristine biomass, the maximum adsorption capacity of the surface-modified biomass for AO8, AB45 and RO16 increased from 33, 18 and 25 mg/g, respectively, to 352, 196 and 338, mg/g (Low et al., 2008).

Biological Processes

Bacterial Processes. A new dye-degrading strain, *Bacillus fusiformis*, was characterized and identified by Kolekar et al. (2008) from an effluent contaminated site of textile dyeing industry. The strain showed the ability to tolerate and degrade azo dyes, Disperse Blue 79 (DB79) and Acid Orange 10 (AO10) under anoxic conditions. Complete mineralization of DB79 and AO10 at the concentration of 1.5 g/L was observed within 48 h.

A consortium-GB containing two microorganisms, *Galactomyces geotrichum* MTCC 1360 and *Bacillus* species VUS, was able to degrade sulfur-containing dye Brilliant Blue G (BBG), optimally at pH 19 and temperature at 50°C. Malt extract, peptone and beef

extract were found to be the best carbon and nitrogen sources. The analysis of degradation products showed formation of new and different products by consortium and pure cultures, when incubated with BB G for 5 h (Jadhav et al., 2008). According to Jadhav et al. (2008) *Galactomyces geotrichum* MTCC 1360 was able to achieve 100% decolorization of a toxic azo dye Methyl Red (100 mg/L) within 1 h in deionized water at 30°C. The degradation was possible in a wide range of pH values (3-12) and temperature (5-50°C). Addition of 1 g/L molasses in deionized water made decolorization possible in only 10 min. Carvalho et al. (2008) assessed biodecolourisation of an azo dye by anaerobic cultures using liposomal textile levelling agent as primary substrate. Liposomes seemed to facilitate the uptake of the dye Acid Orange 7 (AO7) by anaerobic biomass leading to a fast decolourisation. By contrast, the presence of dye (60-300 mg/L) caused a decrease in the COD degradation rate (4.1-2.5 g COD removed /L•d for 60 and 300 mg/L of dye, respectively).

A novel bacterial consortium (TJ-1) which could decolorize Acid Orange 7 (AO7) and many other azo dyes, was developed by Joshi et al. (2008) In TJ-1 three bacterial strains were identified as *Aeromonas caviae*, *Proteus mirabilis* and *Rhodococcus globerulus* by 16S rRNA gene sequence. AO7 degradation was observed under microaerophilic conditions in the presence of organic carbon source. Either yeast extract (YE) alone or a combination of YE and glucose resulted in higher decolorization. More than 90% decolorization was achieved eve at 200 mg/L within 16 h. Deng et al. (2008) isolated and characterized a new strain of *Bacillus cereus*,

DC11), capable of decolorizing a broad-spectrum of dyes. High decolorization efficiency (95-98%) was obtained within 6 h of incubation for 100 μM Acid Blue 25 (anthraquinone dye), 4 h for 55 μM Malachite Green (thiphenylmethane dye), and 2 h for 750 μM Basic Blue X-GRRL (azo dye) at 20-45°C and neutral pH under anaerobic conditions.

A microbiological evaluation was performed by Lima et al. (2008) on two Brazilian treatment plants, one in the polyester dyeing unit wastewater (25 m^3/h), and the other one in the wastewater from a cotton laundry and finishing. (4 m^3/h). Qualitative and quantitative evaluations of the present microfauna in the aeration tank were correlated with the typical parameters of operational control and performance. The results have confirmed the existence of specific microfauna for each type of textile effluent showing also a significant change in the relative percentage of the different groups of microorganisms during the acclimatization of the sludge.

Eight different commonly employed textile dye auxiliaries, namely two dye carriers (Carrier A: a mixture of isobutanol and tetra propylene benzene sulphonate; Carrier B: a mixture of butyl benzoate and dodecyl benzene), two biocidal finishing agents (Biocide A: a 2,4,4'-trichloro-2'-hydroxydiphenyl ether; Biocide B: a nonionic diphenyl alkane derivative), two tannin formulations (Tannin A: a condensation product of aryl sulphonate; Tannin B: tannic acid), and two dispersing agents (Dispersing Agent A: a naphthalene sulphonic acid derivative; Dispersing Agent B: a lignin sulphonate derivative) were investigated in terms of toxicity and

recalcitrance in a comparative manner. The acute toxicity of dye auxiliaries was assessed by employing either *Daphnia magna* (water flea) or *Phaedactylum tricornutum* (marine microalgae). The results related to the activated sludge inhibition test were presented (Germirli Babuna et al., 2008).

Sludge sample from biological treatment plant of a textile industry was acclimatized for decolourisation of azo dye Direct Black 38 (DB38). A continuous culture experiment showed that the acclimatized sludge could decolourize 76% of 100 mg/L DB38 (Bafana et al., 2008). A novel bacterial strain capable of decolorizing reactive textile dye Red BLI was isolated by Kalyani et al. (2008) from the soil sample collected from contaminated sites of textile industry. The bacterial isolate was identified as *Pseudomonas* sp. SUK1 and was able to perform Red BLI (50 mg/L) decolorization efficiency of 99.28% within 1 h under static anoxic condition, at pH in the range of 6.5-7.0 and 30°C. Bafana et al. (2008) isolated an *Enterococcus gallinarum* strain from the effluent of the treatment plant of a textile industry based on its ability to decolourize C.I. Direct Black 38 (DB38). The strain was found to decolourize even under aerobic conditions, degrading resulting benzidine by deamination and utilizing the NH_3 released as sole source of nitrogen for growth.

Decolorization and degradation of polyazo dye Direct Black 22 (DB22) was carried out by Mohana et al. (2008) by distillery spent wash degrading mixed bacterial consortium, DMC. Response surface methodology (RSM) involving a central composite design (CCD) in four factors was successfully employed for the study and optimization

of the decolorization process. Under optimized conditions, the consortium was able to decolorize the dye almost completely (>91%) within 12 h. According to Dafale et al. (2008), anaerobic acclimatization of activated sludge from a textile effluent treatment plant to high concentration of Reactive Black 5 (RB5) could effectively decolorize RB5 dye solutions. The strains viz. *Pseudomonas aeruginosa* and *Bacillus circulans* and other unidentified laboratory isolates (NAD1 and NAD2) were predominantly present in the microbial consortium. The optimum inoculum concentration for maximum decolorization was found to be 1-5 mL of 1800±50 mg/L Mixed Liquor Suspended Solids (MLSS) and 37°C. The biomass showed efficient decolorization even in the presence of 10% NaCl, as tested with RB5. Hong et al. (2008) refer that four strains of dissimilatory azoreducing bacteria (DARBs), isolated from activated sludge of a textile-printing wastewater treatment plant, could reduce azo compound by coupling oxidation of several electron donors.

Russo et al. (2008) assessed two enzyme-based process for the conversion of the anthraquinone dye Remazol Brilliant Blue R. A crude laccase mixture from *Pleurotus ostreatus* was immobilised on EUPERGIT C 250L$^©$, an acrylic resin with epoxy functionalities. Two different fixed bed were used to assay the solid biocatalyst activity and to characterize the dye conversion kinetics. The kinetics of dye conversion were in line with a product-inhibited kinetic model. The kinetics model were implemented to describe dye conversions in a continuous fixed bed reactor (CFBR), operated with immobilised laccase, and in a stirred tank reactor (STR), operated with

free laccases. The CFBR option enabled the remediation of a wastewater volume larger than the one processed in the STR under comparable conditions.

You et al. (2008) studied the performance of sequencing batch reactor (SBR), aerobic membrane reactor (AMBR), anaerobic-oxic membrane bioreactor (AOMBR) and AOMBR/reverse osmosis (AOMBR/RO) to treat a synthetic textile wastewater containing Reactive Black 5 (RB5). After 2 years process operation, the average Suspended Solids (SS) and Biochemical Oxygen Demand (BOD) concentration of the effluent showed that membrane processes did perform an excellent removal efficiency. In addition, it was found that the anaerobic tank of AOMBR and AOMBR/RO could lower the COD concentration. The study also isolated higher decolorization performance microorganisms. An up-flow biological aerated filter packed with two layers media was used by Liu et al. (2008) for tertiary treatment of textile wastewater secondary effluent. Good performance of the reactor were achieved and the average COD, NH_4^+-N and Total Nitrogen in the effluent were 31, 2 and 8 mg/L, respectively. The dissolved oxygen (DO) concentration showed to play a crucial role in substrate removal: the increase of DO concentrations resulted in higher performances for carbon and ammonia removal.

A sequential anaerobic/aerobic reactor system was used by Işik and Sponza (2008) to treat a simulated textile wastewater containing azo dyes (mixing of 50 mg/L of Reactive Black 5, 50 mg/L of Direct Red 28, 50 mg/L of Direct Black 38, 50 mg/L Direct Brown 2 and 50 mg/L of Direct Yellow 12 as 250 mg/L of total concentration),

soluble starch, carboxymethyl cellulose (CMC), acetic acid, glucose, salts, acids and other additives. COD and color removal efficiencies between 87 and 91% and between 84 and 91% were obtained at a total HRT of 19.17 and 1.22 d, respectively.

According to Somasiri et al. (2008), an upflow anaerobic sludge blanket (UASB) treating a real textile wastewater (RTW) showed an efficiency over 90% in reducing COD. The activities of the anaerobic granules were not affected by the recalcitrant nature and high pH of the RTW. Bulc and Ojstrşek (2008) examined the treatment efficiency of constructed wetlands (CW) for the dye-rich textile wastewater with special focus on color reduction. The average treatment efficiency of the CW were: 84% COD, 66 BOD_5, 89% TOC, 87% N_{tot}, 88% sulphate, 80% anionic surfactants, 93% total suspended solids (TSS) and 90% color. Sandhya et al. (2008) showed effectiveness of a microaerophilic–aerobic hybrid reactor in the treatment of textile wastewater. COD and color were reduced to 82-94% and 99%, respectively. When operated at the highest loading of 16.4 g COD/L•d, the reactor obtained 80% COD and 72% color removal.

Fungal Processes. Dyes belonging to the mono-, di-, tri- and poly-azo as well as anthraquinone and mono-azo Cr-complexed classes, were employed by Ciullini et al. (2008) for a comparative enzymatic decolorization study using the extracellular crude culture extracts from the white rot fungus *Funalia* (*Trametes*) *trogii* grown on different culture media. Besides, activators were used able to trigger different levels of expression of oxidising enzymes: laccase and cellobiose dehydrogenase. Laccase containing extracts

were capable to decolorize some dyes from all different classes analysed, whereas the recalcitrant dyes were subjected to the combined action of laccase and the chemical mediator HBT, or laccase plus cellobiose dehydrogenase.

A total of 37 strains of aquatic hyphomycetes and 95 fungal isolates derived from diverse freshwater environments were screened on agar plates for the decolorization of the disazo dye Reactive Black 5 (RB5) and the anthraquinone dye Reactive Blue 19 (RB19). The decolorization of 9 azo and 3 anthraquinone dyes by 9 selected aquatic fungi was subsequently assessed in liquid test system. The majority of dyes were decolorized by several mitosporic aquatic isolates at rates essentially comparable to those observed with the most efficient white rot fungus (Junghanns et al., 2008). The ability of two new white rot fungi *Scizophyllum commune* IBL-06 and *Ganoderma lucidum* IBL-05 to decolorize direct dye Solar Golden Yellow R (SGY) was demonstrated by Asgher et al. (2008) using Kirk's basal salts medium. In initial time course study, the maximum decolorization (73%) of SGY was caused by *S. commune* IBL-06 after 6 d incubation at pH 4.5 and 35°C. Supplementation of the medium with additional carbon sources enhanced dye decolorization. Addition of glucose gave the best results: complete decolorization was achieved after only 2 d of incubation.

Combined Processes

The granular activated carbon (GAC)-Sequencing Batch Reactor (SBR) system was applied by Sirianuntapiboon and Sansak (2008) to treat both synthetic and raw textile wastewater (STWW and TWW, respectively) containing direct dyes (Direct Blue 201 and Direct Red 23) under various concentrations of biomass and dyestuffs. The optimal operating conditions were found to be 3000 mg/L of Mixed Liquor Suspended Solids (MLSS), Hydraulic Retention Time (HR) of 7.5 d and 40 mg/L of direct dyes. The highest removal efficiency was obtained with STWW. By contrast, the system with TWW showed quite low BOD_5 and dye removal efficiencies; however, its efficiency could be increased by adding carbon sources.

A laboratory-scale biological activated carbon (BAC) process was used to treat a reactive azo dye (Reactive Red 22, RR22) by *Pseudomonas luteola*. The BAC-reactor removed 89% of RR22 while *P. luteola* and suspended *P. luteola* reached a maximum growth rate at a steady-state condition. The *P. luteola* biofilm bioregenerated the activated carbon by lowering the RR22 concentration at the biofilm/activated carbon interface (Lin and Leu, 2008). Mezohegyi et al. (2008) studied the anaerobic decolourisation of azo dye Acid Orange 7 (AO7) in a continuous upflow stirred packed-bed reactor (USPBR) filled with biological activated carbon (BAC). The application of appropriate stirring in the carbon bed resulted in an increase of azo dye bioconversion up to 96% in 0.5 min, compared to unstirred reactor.

The decolorization of aqueous solutions Methylene Blue (C.I. Basic Blue 9), due to the presence of nanocrystalline Bi_2S_3, supported on SiO_2 submicron particles, was investigated by Albuquerque et al. (2008). For this decolorization process, two distinct characteristics,

though related, associated to the role of Bi_2S_3/SiO_2 were identified: (1) high MB adsorption capability and (2) photocatalytic activity to MB photodecolorization. The maximum adsorption ability of the Bi_2S_3/SiO_2 was approximately 15.6 mg MB per gram. The complete decolorization of the dye was obtained with both mechanisms.

Wu (2008) considered the effects of ultrasound US) irradiation on the decolorization of C.I. Reactive Red 198 (RR198) in UV/ZnO system. It was observed that the decolorization rate of RR198 increased with the ZnO dosage in 0.1-1 g/L and with pH in the UV/ZnO system. At pH 7, the decolorization rate constants of UV/US/ZnO, UV/ZnO, US/ZnO, UV/US and US were 0.0739, 0.0534, 0.0022, 0.0020 and 0.0013 min^{-1}, respectively. The results obtained indicated that the UV/US/ZnO system was able not only to completely decolorize RR198 but also to effectively mineralize the dye.

Degradation of Rhodamine B (RB) and COD by means of the coupled photocatalytic oxidation (PCO) and biofilm systems was studied by Kim and Park (2008). The coupled PCO-biofilm systems were divided into two operation par: the R1 consisted of a pre-PCO and a post-biofilm reactor system, while the R2 was a pre-biofilm and a post-PCO reactor system. In a batch experiment, the order of initial decolorization was photocatalytic oxidation>adsorption>photolysis. In a continuous experimtal study, almost all color was removed in the PCO reactor. During 180 d, color and COD removal efficiencies in the R2 system were higher than those in the R1 system.

The commercial strong-acid cation exchange membranes (either with sulfonic groups or phosphate groups) could successfully remove cationic dye Methyl Violet 2B (MV) from water and allowed almost complete dye recovery under appropriate desorption condition (1 M NaCl 60% methanol). The performance of both membranes remained practically unaltered during three successive cycles of adsorption and desorption (Wu et al., 2008). Catalytic thermal treatment (thermolysis) accompanied with coagulation was used by Kumar et al. (2008) for the removal of COD and color of composite wastewater from a cotton textile mill. Homogeneous copper sulfate at a mass loading of 6 kg/m^3 was found to be the most active catalytic agents. Similarly, during coagulation aluminum potassium sulphate [$KAl(SO_4)_2 \cdot 16H_2O$] at a coagulant concentration of 5 kg/m^3 was found to be the best coagulants tested. During thermolysis, a reduction of COD and color of about 77.9% and 92.9%, respectively, was observed at pH 12. Coagulation of the supernatant obtained after thermolysis resulted in overall reduction of 97.3% COD and close to 100% color at pH 8.

Sahinkaya et al. (2008) applied the activated sludge (AS) treatment coupled with nanofiltration to improve denim textile wastewater quality to reuse criteria. In the AS reactor, the COD removal efficiency was quite high, about 90% and 84%, on the basis of total and soluble COD, respectively. The color reduction was in the range of 65-85% and 50-70% of removed color was adsorbed on biomass or precipitated within the reactor. Although the high performances of AS, the effluent did not meet criteria for reuse and the NF was applied as a further treatment.

Besides, microfiltration was added before NF to remove coarse particles. NF system allowed almost complete rejection of color and also high rejection of COD was observed.

Photocatalysis was able to remove more than 90% color from crude as well as autoxidized chemically reduced dye solutions representative of recalcitrant dye residues after biological sequential anaerobic-aerobic treatment. The end products of photocatalytic treatment were not toxic toward methanogenic bacteria. Therefore, it was suggested that the photocatalysis can be used as a pre- or post-treatment to biological anaerobic process for textile wastewater (Harrelkas et al., 2008). A 20 L external-loop airlift reactor was used by Essadaki et al. (2008) as an electrochemical cell in order to carry out water depollution using batch electrocoagulation (EC) without mechanical agitation, pumping requirements or air injection. Mixing and complete flotation of the pollutants were achieved using only the overall liquid recirculation induced by H_2 microbubbles generated by water electrolysis. The innovative application of airlift reactors was used for decolorization of a red dye. Operation time and removal efficiencies were similar to those reported in conventional EC cells, but specific energy and electrode consumptions were smaller. Yanhe et al. (2008) explored the feasibility of the combination of enhanced adsorption for Acid Orange 7 (AO7) in solution and regeneration of activated carbons fibers (ACFs) by electrochemical method. The results showed that the adsorption rate and capacity at the polarization of 600 mV increased by 120% and 115%,

respectively. Regeneration efficiency of over 90% was achievable at −5V.

Various

The degradation of Orange G, a mono azo dye, in aqueous solutions was investigated by Bokare et al. (2008) using Fe-Ni bimetallic nanoparticles. Batch experiments with a minimum nanocatalyst loading of 3 g/L showed complete dye degradation after 10 min of reaction time. The degradation efficiency was linearly dependent on the initial dye concentration, pH of the solution and total Fe-Ni catalyst concentration. A study was conducted by Zhuo et al. (2008) with FeO_x-MoO_3-P_2O_5 prepared by solid reaction method and employed as catalysts for degradation of Methylene Blue at mild conditions. The orthogonal experiments were devised to determine the optimum operating conditions. It was possible to achieve 98% of color and 85% of COD reduction in 30 min by adding 13.3 g/L of FeO_x-MoO_3-P_2O_5 catalyst with 25 wt% starch addition.

The degradation of the dye Alizarin Red (AR) was carried out in the hybrid gas-liquid dielectric barrier discharge (DBD) plasma. Based on the bond dissociation energies (BDEs) theory and experimental results, the degradation pathway of AR in DBD plasma was theoretically proposed. It was found that the degradation course consisted of three steps in the order to gas-discharge phase, ring-open phase, and mineralization phase (Xue et al., 2008). A non-thermal plasma technique (i.e., the gas-liquid gliding arc discharge, which is generated between at least two metal electrodes with high voltage) was used by

Du et al. (2008) for the decolorization of 180 μM aqueous solutions of Acid Orange 7 (AO7). The concentration of AO7 solution decreased exponentially to reach 58.9, 77.4, 89.1, 95.1 and 99% in 25, 50, 75, 100 and 125 min, respectively.

Ting and Jamaludin (2008) studied the effect of the concentration of pollutants to their removal using electron beam technology. Irradiation was found to be effective in removing highly colored and refractory organic compounds. The color removal for initial concentrations of 255 CU, 520 CU, 990 CU and 1900 CU treated using irradiation at 0.5 kGy were 61, 48, 28 and 16%, respectively. COD removal showed similar trend but at relatively lower removal percentage.

According to Peng et al. (2008), a combination of ferric chloride and sodium nitrite significantly improved the wet oxidation of the azo dye Acid Orange 7 (AO7) in acid aqueous medium (pH 2.6) under moderate conditions (T=150°C; oxygen pressure=0.5 Mpa). At these conditions, 56% TOC was removed after 4 h treatment, while no obvious TOC removal was achieved without catalyst at the same experimental conditions. The catalytic performance of 3 wt% copper supported on carbon nanofibers (CNFs) in liquid phase oxidation was tested by Rodríguez et al. (2008) using a batch stirred microreactor in order to determine the decolorization and TOC removal efficiency in washing textile wastewater (WTW). More than 97% color reduction and 81% TOC removal were obtained by properly choosing the operation conditions after 180 min of reaction.

Zhang et al. (2008) studied the synergistic effects of microwave (MW) irradiation induced oxidation processes and $CoFe_2O_4$ for the degradation of Brilliant Green (BG) from aqueous solutions. Under the optimum conditions, the ratio of solid to liquid ratio was 1:167 (0.3 g $CoFe_2O_4$ mixed with 50 mL of 20 mg/L BG solution), MW power of 600 W, and the irradiation time of 2 min. Under these conditions, decolorization reached 100%.

The potential use of calixarenes as low-cost efficient extractant for the removal of azo dyes from wastewater was studied by Gungor et al. (2008). The carboxylic acid derivative of calyx[8]arene showed highest affinity towards the azo dyes. The extent of the dye removal increased with the addition of NaCl.

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