

## CELLULOSIC SUBSTRATES FOR REMOVAL OF POLLUTANTS FROM AQUEOUS SYSTEMS: A REVIEW. 2. DYES

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Dyes used in the coloration of textiles, paper, and other products are highly visible, sometimes toxic, and sometimes resistant to biological breakdown; thus it is important to minimize their release into aqueous environments. This review article considers how biosorption of dyes onto cellulose-related materials has the potential to address such concerns. Numerous publications have described how a variety of biomass-derived substrates can be used to absorb different classes of dyestuff from dilute aqueous solutions. Progress also has been achieved in understanding the thermodynamics, kinetics, and chemical factors that control the uptake of dyes. Important questions remain to be more fully investigated, such as those involving the full life-cycle of cellulosic substrates that are used for the collection of dyes. Also, more work needs to be done in order to establish whether biosorption should be implemented as a separate unit operation, or whether it ought to be integrated with other water treatment technologies, including the enzymatic breakdown of chromophores.

**Keywords:** Cellulose; Biomass; Biosorption; Remediation; Pollutants; Adsorption; Textile dyes; Basic dyes; Direct dyes; Reactive dyes; Wastewater treatment

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### INTRODUCTION

The visibility, toxicity, and persistent nature of various synthetic dyestuffs have provided motivation for an extensive body of research aimed at preventing their release to natural environments. This article reviews studies whose goal has been to remove dyes from aqueous solution by their adsorption onto various cellulosic or cellulose-derived materials. In this article the term “dyes” will be used in a broad sense to include any colorant used in production of textiles, paper, and various other manufactured items.

The term “biosorption” is widely used to denote the practice of using plant-derived matter, either dead or alive, to remove unwanted soluble materials from water (Gadd 2009; Park *et al.* 2010). Several review articles have dealt with aspects of the biosorption of dyes (Sanghi and Bhattacharya 2002; Pearce *et al.* 2003; Aksu 2005; Crini 2006; Hashem *et al.* 2007; Demirbas 2009; Gupta and Suhas 2009; Foo and Hameed 2010; Rafatullah *et al.* 2010; Srinivasan and Viraraghavan 2010; Sharma *et al.* 2011a). In addition, various authors have reviewed a closely related topic, biodegradation of dyes,

in which enzymes facilitate molecular decomposition and decolorization (Banat *et al.* 1996; Fu and Viraraghavan 2001a; Husain 2006, 2010). The present article attempts to achieve a more comprehensive view of biosorption of dyes by drawing upon a greater proportion of the available literature, including more recent articles.

The potential discharge of dyestuffs to the environment worldwide has been estimated to be of the order of magnitude of  $7 \times 10^4$  tons per year (Vijaraghavan and Yun 2008b). Release of dyes to the environment is often associated with industrial plants (see, for instance O'Neill *et al.* 1999; Chakraborty 2010; Malik and Bharti 2010). Conventional wastewater treatment technologies, which will be discussed in the next section, have been only partly successful in removing all such materials from effluent streams (Slokar and Le Marechal 1997; Vandevivere *et al.* 1998; Robinson *et al.* 2001; Forgacs *et al.* 2004; Joshi *et al.* 2004; Mondal 2008). Problems arise due to the diverse nature of commonly used dyes, their solubility in water, and the fact that they are designed to be highly visible. Many modern dyestuffs have a high resistance to biological decay (Aksu 2005), so they are only partly removed from water during conventional secondary wastewater treatment, *i.e.* with the use of activated sludge.

## OVERVIEW OF DYE REMEDIATION TECHNOLOGIES

To provide context for the main topic of this article – the biosorption of dyes – the present section reviews alternative technologies, including those that are in most common use. Though the currently used technologies for water treatment are highly effective for removal of conventional pollutants and toxicity (Wiesmann 2007), they are not particularly effective for removal of dye molecules and their associated color from wastewater (Horning 1978; Tunay *et al.* 1996; Hao *et al.* 2000; Forgacs *et al.* 2004; Singh and Arora 2011). Thus, a treated discharge may be very low with respect to typical measures of pollution such as biochemical oxygen demand (BOD), chemical oxygen demand (COD), total suspended solids (TSS), metals, and aquatic toxicity, but still be objectionable due to the color imparted to the receiving stream. As noted by Huber and Carré (2012), rhodamine dye is particularly recalcitrant, such that effluents obtained from the deinking of paper often have a red coloration, even after conventional wastewater treatment. Color discharges may also accumulate in sediments, impair naturally occurring processes that require light penetration, and limit downstream beneficial uses such as drinking water, irrigation, and recreation. Even in the absence of adverse environmental impacts, the presence of color in receiving streams may cause public concerns, resulting in negative relationships within the community and leading to regulatory action. Regulations and international standards such as those set forth by the BSR Sustainable Water Group and the World Health Organization establish limits requiring that discharges be “free from” color or meet specific color limits. For these reasons, methods of color removal have long been studied and implemented in wastewater treatment processes.

Comprehensive summaries of dye decolorization processes have been compiled from the literature by several authors (Tunay *et al.* 1996; Slokar and Marechal 1998; Hao *et al.* 2000; Robinson *et al.* 2001; Rai *et al.* 2005; Latif *et al.* 2010; Saratale *et al.* 2011;

Siddique *et al.* 2011; Singh and Arora 2011; Verma *et al.* 2012; Huber and Carré 2012). The wastewater from dyeing operations can be expected to contain not only mixtures of different dyestuffs, but also a variety of dispersants, inorganic ions, and diverse pH (Correia *et al.* 1994; O'Neill *et al.* 1999). The processes most widely studied and used in full scale operations are briefly discussed below.

The discharge of oxygen-demanding substances found in textile wastewater to the receiving stream is of primary concern. The oxidation of these substances by naturally occurring bacteria and other organisms can result in the depletion of oxygen in the receiving stream, creating septic conditions and adversely impacting aquatic life. Therefore, implementation of treatment technologies that reduce the oxygen demand, as measured by BOD and COD, prior to discharge, is a first priority. The most widely used treatment technology for this purpose is the activated sludge process (Wiesmann 2007). In this process, bacteria are used to oxidize organics prior to discharge. The biological process requires large amounts of oxygen and results in the production of biological sludge due to bacterial growth. The sludge is typically separated by sedimentation processes, dewatered, and disposed by means of land application for agricultural uses or in landfills. Though activated sludge treatment is essential and is the main treatment process used for textile wastewater treatment, its ability to remove color is limited and dependent on the types of dyes found in the wastewater.

In general, textile dyes are resistant to aerobic biological treatment, and minimal removal due to bio-oxidation occurs (Rai *et al.* 2005). However, removal has been observed through adsorption onto the activated sludge biomass (Porter and Snider 1976; Pagga and Taeger 1994). Thus, operation of biological systems at high sludge growth rates (low sludge age) should result in higher rates of color removal (Davis *et al.* 1982). Dyes adsorbed to the sludge biomass are then disposed with the sludge. However, even under optimal growth conditions, adequate color removal is rarely achieved by aerobic biological treatment, and additional treatment usually is required.

Anoxic biological treatment processes have been shown to decolorize certain azo reactive dyes (Smith *et al.* 2007). Under anaerobic or anoxic conditions, the nitrogen-nitrogen azo bond is cleaved and the dye chromophore is destroyed. Though this reaction provides decolorization, it does not result in the destruction of organics, and dye molecule fragments persist. Thus, anoxic and anaerobic processes are typically followed by aerobic treatment for oxidation of remaining organics.

The decolorization process most widely used in conjunction with biological treatment is chemical coagulation (Verma *et al.* 2012). Aluminum salts such as alum and polyaluminum chloride (PAC), iron salts such as ferric chloride and ferrous sulfate, and organic coagulants such as polyamines have been found to be effective for removal of some types of dyes. Disperse, vat, and sulfur dyes can generally be effectively precipitated through such chemical coagulation (Horning 1978). Removal by chemical coagulation of highly water-soluble dyes such as reactive dyes is less effective. In addition to the cost of chemicals, chemical coagulation results in additional production of sludge, which then must be removed and dealt with appropriately.

Electrochemical processes that generate iron or aluminum ions through electrolysis have been studied (Lin and Peng 1994; Naumczyk *et al.* 1996). A combination of reactions occurs simultaneously during this process. Oxidation occurs as a result

of chlorine and ozone gas production at the anode. Hydrogen gas produced at the cathode may aid in the flotation and separation of coagulated solids. Iron or alum “sacrificial” anodes release ions to the liquid and act as coagulating agents. These processes are effective for dyes amenable to chemical coagulation as well as reactive dyes that are amenable to oxidation. The use of electrochemical processes is limited due to the high capital and operating cost as well as the cost for disposal of sludges produced.

Oxidation processes, including ozonation (Sarasa *et al.* 1998) and hyper-chlorination, may be effective for certain soluble dyes such as reactive dyes (Churchley 1994; Liakou *et al.* 1997a,b; Arslan-Alaton 2003). Chlorine is more effective at low pH. However, the use of chlorine for color removal is not encouraged due to concerns regarding the formation of chlorinated by-products including trihalomethanes. Ozone may also produce by-products that require further oxidation and result in the release of metals from the dye structure (Adams *et al.* 1995). High capital costs are associated with ozonation systems; however, operating costs may be favorable, depending on the cost of electricity.

Advanced oxidation processes involve the generation of highly reactive free radicals, specifically the hydroxyl radical, for oxidation of the dye molecule. UV light is generally used to generate the free radical, reacting with hydrogen peroxide ( $\text{H}_2\text{O}_2$ ) or ozone ( $\text{O}_3$ ). In some studies, titanium dioxide ( $\text{TiO}_2$ ) has been used as a catalyst (Li and Zhang 1996). Some reactive, basic, acid, and direct dyes have been shown to be readily decolorized, while dispersed and vat dyes were only partially decolorized (Yang *et al.* 1998). Though effective, advanced oxidation processes have not been widely used in full-scale applications due to the high cost of chemical and ultraviolet systems.

Membrane processes can be very effective for removal of most dyes (Fersi *et al.* 2005; Jirankova *et al.* 2010; Latif *et al.* 2010). Membranes can be selected based on their pore size and their ability to nominally retain solutes at a specified molecular weight cutoff (MWCO) measured in Daltons. The MWCO range for nanofiltration is between 300 and 1000, and such membranes are likely to be the most suitable selection for rejection of dissolved organics, including dyes. The operating pressure and operating costs generally increase as the MWCO is decreased due to increased energy and membrane maintenance costs. Membrane treatment does not destroy, but rather retains color in a concentrated wastestream. Therefore, relatively high volumes of concentrated color are collected that require additional treatment. Due to the high capital and operating cost of membrane systems and the additional cost for treatment or disposal of the concentrate, membrane systems are not generally used for wastewater treatment. Membrane systems may be cost-effective for small-scale water reuse systems. If a means of disposal of the concentrate without significant treatment is available, the cost effectiveness of these systems is greatly improved. Different types of membrane processes were evaluated by Fersi *et al.* (2005) for the treatment of biologically treated effluent. They found that nanofiltration produced permeate suitable for reuse in the dyeing process.

Considerable study of the use of white-rot fungi for decolorization of azo dyes has been performed (Wesenberg *et al.* 2003). White-rot fungi (WRF) produce various extracellular enzymes that degrade lignin in their natural lignocellulosic environment. The primary enzymes involved in the breakdown of lignin are laccase, manganese

peroxidase, and lignin peroxidase. These oxidases have also been found to degrade various xenobiotic compounds including dyes. Work performed by Swamy and Ramsay (1999) evaluated five species of WRF for their ability to degrade five different azo dyes. They found that *Tinea versicolor* demonstrated the greatest degree of decolorization and rate of decolorization of the species tested. Further work is needed to identify the degradation pathway and the end products from enzymatic degradation of dyes.

While this and other work show the potential for enzymatic degradation of dyes, several obstacles must be overcome for commercialization. The major challenge is development of a suitable reactor and operating conditions for full-scale treatment. The impact of other contaminants in effluents that may interfere with enzyme degradation must also be evaluated.

The adsorption of color onto various substrates has been extensively studied. As with other technologies, all dyes are not effectively adsorbed, and loadings vary from one substrate to another. Reactive, basic, acid, and azoic dyes can be readily removed using activated carbon (Horning 1978; Foo and Hameed 2010). However, carbon systems are not widely used due to the high cost associated with carbon replacement or regeneration. Adsorption efficiency is affected by solution pH. The presence of other organic and inorganic molecules that compete for adsorption sites may reduce carbon loadings and further increase operating costs. As will be discussed in more detail in later sections, other substrates have been studied, such as chitosan fiber (Yoshida *et al.* 1991; Yoshida and Takemori 1997), wood chips (Poots and McKay 1976), fly ash (Ramakrishna and Viraraghavan 1997), bagasse (McKay *et al.* 1987), rice husks, and other biomaterials (Laszlo 1994). These materials have demonstrated adsorption affinity for various dyes but may be selective for a specific type of dye class. Problems associated with material handling, separation, and disposal of these materials have generally discouraged their use on a large scale. The economic availability of these materials is often based on their proximity to the application, which may further limit their use.

In principle it would be feasible to adsorb dye molecules at industrial scale onto the surfaces of lignocellulosic sorbent materials that are in suspension in a batch reactor. According to Volesky (1987), such systems tend to have high costs for both construction and operation. For practical applications of biosorption, continuous processes involving flow-through columns have been widely studied (Robinson *et al.* 2002a; Aksu *et al.* 2007a,b; Han *et al.* 2007a; Padmesh *et al.* 2006a,b; Li and Jia 2008; Ramasamy *et al.* 2008; Vijayaraghavan *et al.* 2008b; Saeed *et al.* 2009). Bed-type systems can be expected to have practical advantages such as reduced space requirements and the fact that they incorporate a means to separate the biomaterial from the aqueous phase to be discharged. In particular, they avoid the need to implement a separate process to collect the biosorbent from suspension after it has been used in a batch-type operation (Vijayaraghavan and Yun 2008b). On the other hand, column-type separation systems can place severe limitations on the physical form and mechanical strength of the sorbent material; biosorbents that are too deformable can be expected to become compressed, thus tending to seal off flow in a column (Vijayaraghavan and Yun 2007a). Such effects are especially of concern when considering the possible use of microbial biomass (Vijayaraghavan and Yun 2008b). Though an up-flow mode of column use has been suggested (Vijayaraghavan and Yun 2008b), presumably as a way to minimize such

issues, a more effective way to achieve good packing and flow characteristics in a flow-through column involves immobilization of the fine compressible material on the surfaces of other solid material, which may be lignocellulosic. A further advantage of immobilizing finely divided biosorbents onto more rigid, highly permeable media is to minimize the plugging of the bed by migration of fine particles to locations where channels in the bed become plugged (Hubbe *et al.* 2009).

Insights into the practical challenges in using biosorption to purify dye-containing wastewater can be gained by considering the fact that the amount of dye adsorbed tends to increase with increasing concentration in solution. However, there can be strict limits on the final concentration of dye in solution at the end of the water treatment process. This implies that a single-stage treatment, based only on biosorption, could require an extremely large mass of material in proportion to the amount of dye that needs to be removed. The ideal system would allow near-quantitative adsorption at ratios of dye to sorbent corresponding to the initial color concentration, which would greatly reduce the mass of adsorbent required. Thus, the application of adsorption systems may require the use of counterflow systems, such as columns or multi-stage contactors. This requirement further increases the complexity and capital cost of adsorbent systems.

The effectiveness of color removal technologies for textile wastewaters is dependent on many factors, including the type of dyes present, the concentration of other competing or interfering materials, and process conditions such as pH and temperature. Textile wastewaters generally contain multiple types of dyes, and a combination of removal technologies may be required to meet treatment objectives. Color removal generally requires the use of specific unit operations in addition to processes required for the removal of conventional pollutants. These additional, tertiary processes increase the cost of facilities, cost of operations, and the complexity of operations. Thus, additional research is needed to identify low-cost color removal materials and technologies that can be used for multiple types of dyes. Cellulosic substrates may be suitable for this application, providing a use for waste materials and/or materials obtained from sustainable sources.

Various authors have noted that conventional wastewater treatment approaches can be costly and difficult to implement in the case of small facilities to treat the wide range of dye-containing wastewater from various operations (Culp 1963; Hameed *et al.* 2007; Pavan *et al.* 2008; Pekkuz *et al.* 2008; Kang *et al.* 2009; Raposo *et al.* 2009; Li *et al.* 2010; Sharma *et al.* 2010; Gao *et al.* 2011; Siddique *et al.* 2011). Among the physical processes, adsorption merits particular attention due to its simple handling and cost-effective characteristics. It has potential to be an efficient technique for color removal from wastewater and dyestuff effluents, and the adsorption process can yield high quality treated water. The major advantages of an adsorption system for water pollution control may include a relatively low initial investment, a simple design, easy operation, low energy intensiveness, the use of non-toxic materials, and superior removal of organic waste constituents at low concentrations as compared to the conventional biological treatment processes (Imran and Gupta 2006; Yao *et al.* 2010; Sharma and Uma 2010; Uddin *et al.* 2009).

## DYE CHARACTERISTICS AFFECTING THEIR UPTAKE BY CELLULOSICS

The coloring of textiles with dyes is a process that has been used for 50 centuries. Modern textile dyes contain structural groups that confer color (chromophores), affect hue and solubility (auxochromes), or react with the substrate (Holme 2002). These components may have an influence on the behavior of the dye while it is being applied to the fiber, and here we are especially concerned with their behavior if and when they are present in effluent. Dyes may be classified according to their chemistry, *e.g.*, azo, anthraquinone, triarylmethane, *etc.*, by their end-use, *e.g.*, food, textiles, leather, paper, or by application class, *e.g.* basic, acid, direct, reactive, disperse, *etc.* In this review, dyes will be grouped according to their application class. This is the classification system used by the Society of Dyers and Colourists and the American Association of Textile Chemists and Colorists in the Color Index. Dyes listed in the Color Index may or may not be associated with a known structure, but all listed dyes are identified by a C.I. (Color Index) number, *e.g.*, 111235 for Disperse Yellow 163 (see Table 1).

**Table 1.** Dye Classes and their Properties

Dye Class	Fibers Dyed	Charge	Solubility in Water	Fixation Mechanism	Application Process
Basic/Cationic	Acrylic, modacrylic, cationic dyeable polyester	Positive	Soluble	Ionic bond	Batch
Acid	Nylons, wool, silk, spandex	Negative	Soluble	Ionic bond	Batch
Direct	Cotton, linen, ramie, rayon, lyocel	Negative	Soluble	Hydrogen bond	Batch
Reactive	Cotton, linen, ramie, rayon, lyocel	Negative	Soluble	Covalent bond	Batch or continuous
Disperse	Polyesters, nylons, acetate, triacetate, spandex	Neutral	Very slightly soluble	Dye dissolved in fiber	Batch or continuous
Vat	Cotton, linen, ramie, rayon, lyocel	Neutral	Insoluble	Physical entrapment	Batch or continuous
Sulfur	Cotton, linen, ramie, rayon, lyocel	Neutral	Insoluble	Physical entrapment	Batch or continuous
Naphthol	Cotton, linen, ramie, rayon, lyocel	Neutral	Insoluble	Physical entrapment	Continuous

Dyes may be ionically charged or neutral, and this property certainly affects their removal from a waste stream. The presence or absence of ionic charge also affects the water-solubility of dyes. Table 1 presents a summary of dye classes, their charge, and their solubility in water. The table is organized so that the first four classes of colorants listed are those that have received the most research attention in terms of biosorption.

As indicated in Table 1, dyes are applied to textile fibers, yarns, and fabrics in both batch and continuous processes, in which they are brought into contact with a bath containing the dye and auxiliaries. The dye migrates from the solution to the fiber surface (exhaustion), diffuses from the surface into the fiber, and then is fixed by one of several mechanisms (see Table 1). The fundamentals of the dyeing process have been described in reference works such as those of Needles (1986), Shore (1990), and Hunger (2003). An excellent detailed discussion of textile auxiliaries used in these processes may be found in Shore (2002). The major dyestuff companies also have websites that provide some information about the composition of their products (Clariant, DyStar, Huntsman URL's).

**Table 2.** Types of Auxiliaries Found in Typical Textile Dyebaths

Dye Class	Surfactant	Acid	Alkali/ Base	Electro- lyte	Chelant	Oxidizing Agent	Reducing Agent	Retarder Leveler	Other
Basic/ Cationic	Nonionic wetting agent	Acetic						Anionic or Cationic	
Acid	Anionic wetting agent	Acetic, HCOOH latent, e.g., NH <sub>4</sub> OAc	NH <sub>3</sub>		EDTA or other			Anionic, Cationic or Ampho- teric	
Direct	Anionic wetting agent			NaCl, Na <sub>2</sub> SO <sub>4</sub>					
Reactive	Nonionic or Anionic wetting agents	Acetic	NaOH Na <sub>2</sub> CO <sub>3</sub>	NaCl, Na <sub>2</sub> SO <sub>4</sub>					Cationic fixative
Disperse	Nonionic or Anionic dispersing agents	Acetic	NaOH		EDTA	Sodium m-nitro- benzene sulfonate	Na <sub>2</sub> S <sub>2</sub> O <sub>4</sub>		Anti- migrant
Vat	Anionic dispersing agents		NaOH	NaCl, Na <sub>2</sub> SO <sub>4</sub>		H <sub>2</sub> O <sub>2</sub> NaBrO <sub>3</sub>	Na <sub>2</sub> S <sub>2</sub> O <sub>4</sub>		Anti- migrant
Sulfur	Anionic wetting agent			NaCl, Na <sub>2</sub> SO <sub>4</sub>	EDTA	H <sub>2</sub> O <sub>2</sub> NaBrO <sub>3</sub>	Na <sub>2</sub> S or sugar		Anti- migrant
Naphthol	Anionic wetting agent, Nonionic wetting agent	HCl, Acetic	NaOH, NaOAc	NaCl, Na <sub>2</sub> SO <sub>4</sub> , NaNO <sub>2</sub>	EDTA				Aromat- ic amines, Naph- thols

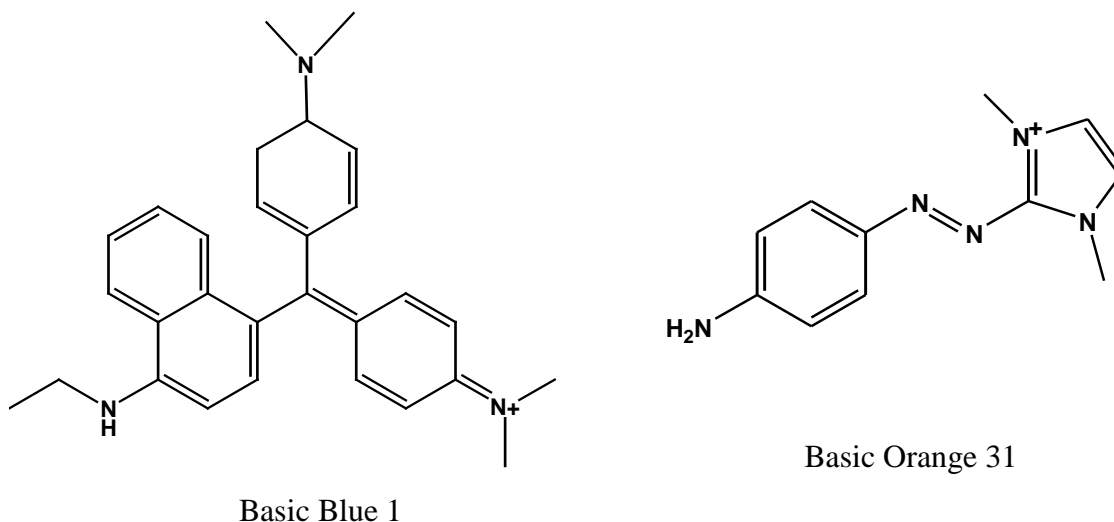


Classes of dyes have been developed to provide coloration and fastness properties for the major types of fibers. The characteristics of the polymer comprising the fiber determines the class of dye used to color it. Structural relationships between the fiber and the dye, processing conditions, and end-use properties are all factors in the selection of auxiliaries that are used in the dye bath. Table 2 presents a summary of types of auxiliaries found in processes used for the various classes of dyes. For each major class of dyes, a brief discussion of the dyeing process and a couple of sample dye structures will be presented.

#### *Basic dyes and their use with acrylic fibers*

Acrylic fibers contain anionic groups, either from the initiator or a co-monomer, and these are used to attach cationic dyes through an ionic bond (Schuler 1980a). Below the glass transition temperature ( $T_g$ ) (72°C, Aitken 1991) of wet acrylic fiber, dye exhaustion is very slow, so heating rates can be relative high in order to compensate. Within the  $T_g$  region, the rate of heating can be manipulated to control the rate at which dyes exhaust to the more accessible regions of the acrylic polymer. In addition to the lowering of heating rates, exhaustion can also be slowed by the addition of retarders. Anionic retarders complex with the cationic dye, and the complex slowly breaks down thermally to release the dye, which then exhausts quickly to the fiber surface. Cationic retarders are typically small alkyl ammonium salts or positively charged polymers that occupy the anionic dye sites on the fiber and are gradually displaced by the dyes, which have greater affinity for the fiber.

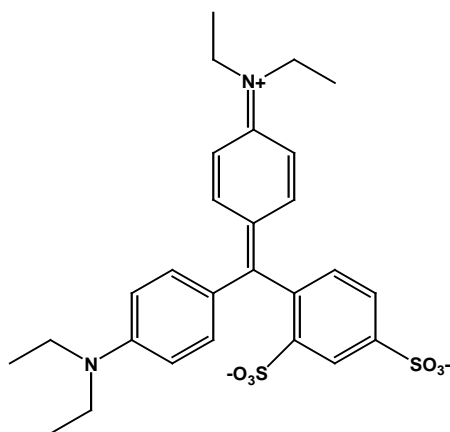
Basic Blue 1 is an example of a triarylmethane cationic dye, and Basic Orange 31 is an example of a cationic azo dye (Fig. 1).



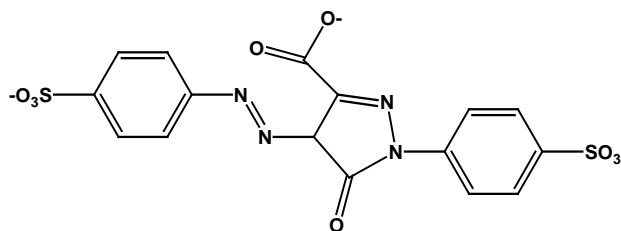
**Fig. 1.** Structures of representative basic dyes, which are often used in coloring of acrylic fiber materials

*Acid dyes and their usage with nylon, wool, and silk fibers*

Acid dyes are relatively small and compact molecules, and they are also anionic, typically as a result of sulfonation. When a polyamide (nylon), wool, or silk is placed in an acidic medium, the basic amino groups are protonated, forming a cationic site to which the anionic acid dye can bind. Because of their structures, wool and silk contain more cationic dye sites than conventional nylon fiber materials. During the dyeing process, the dye exhausts quickly to the cationic fiber surface. To promote level dyeing, the rate of exhaustion is normally controlled by adjusting the pH of the bath with latent acids, controlling the rate of temperature rise, and by the addition of anionic or cationic retarders. Cationic retarders are thought to form thermally labile ionic complexes with the dyes. On heating, these complexes slowly release dye molecules, which then move to the fiber surface in a level manner. Anionic retarders compete with the dye molecules for the cationic dye sites on the fiber surface and are gradually displaced because the dye molecule has a greater affinity than the retarder for the fiber. Acid Blue 45 contains a triaryl methane chromophore, and Acid Yellow 23 is an azo dye (Fig. 2).



Acid Blue 45

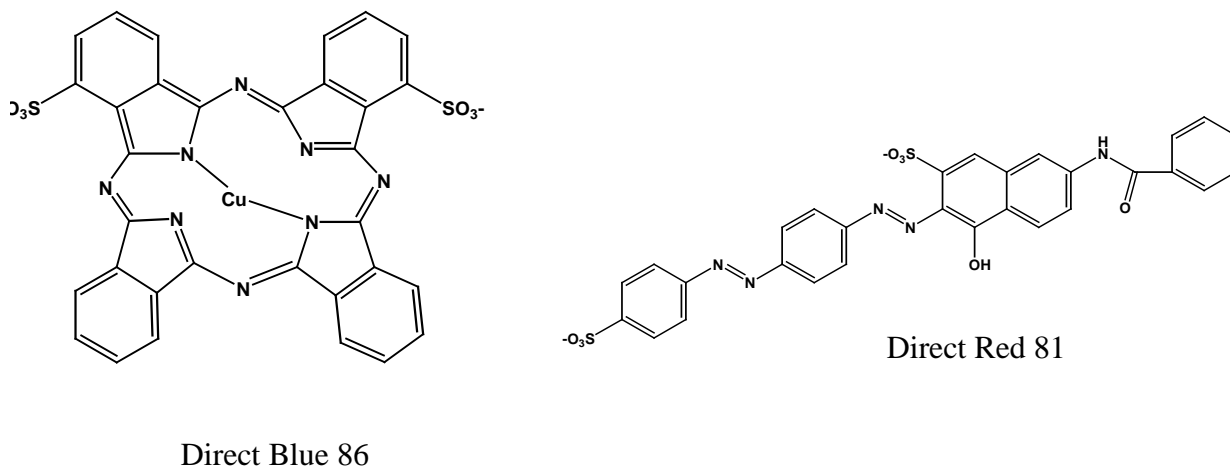


Acid Yellow 23

**Fig. 2.** Examples of acid dyes, which are commonly used for coloring of wool or silk materials, using a low pH of application

*Direct dyes and their use with cellulosic fibers*

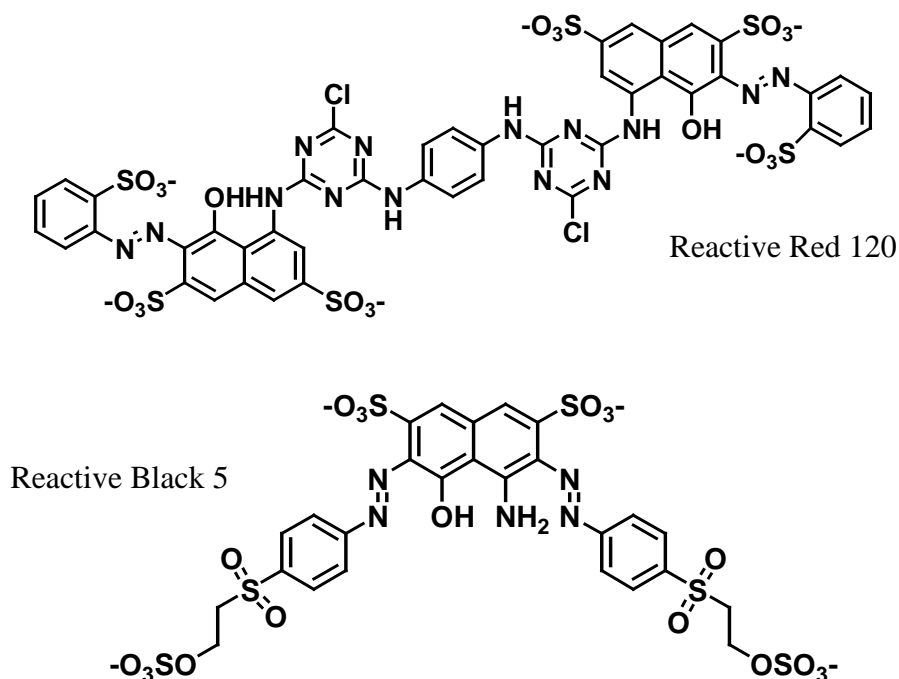
Direct dyes are typically long, linear, anionic, azo dyes that hydrogen bond to cellulosic fibers. In addition to textile applications, direct dyes are also widely used in papermaking (Lips 1981). Subclasses A, B, and C of direct dyes are determined by molecular weight and the factors, *e.g.*, salt and temperature, that are used to control exhaustion. In production, a mixture of dyes will normally include only dyes from one subclass. Sodium chloride and sodium sulfate are the most commonly used electrolytes. Direct Blue 86, a copper-complexed phthalocyanine, and Direct Red 81 are examples of direct dyes (Fig. 3).



**Fig. 3.** Examples of (anionic) direct dyes that are commonly used for coloration of cellulosic fibers both in textile and papermaking applications

#### *Reactive dyes and their use with cellulosic fibers*

The dominant class of textile dyes for coloring cellulosic fibers are reactives. They are water-soluble, bright dyes that react to form a covalent, typically ether bond with the cellulose. To promote the reaction with cellulose, alkali, in the form of sodium hydroxide or sodium carbonate, is added to increase the nucleophilicity of the hydroxyl groups on the anhydroglucose units.

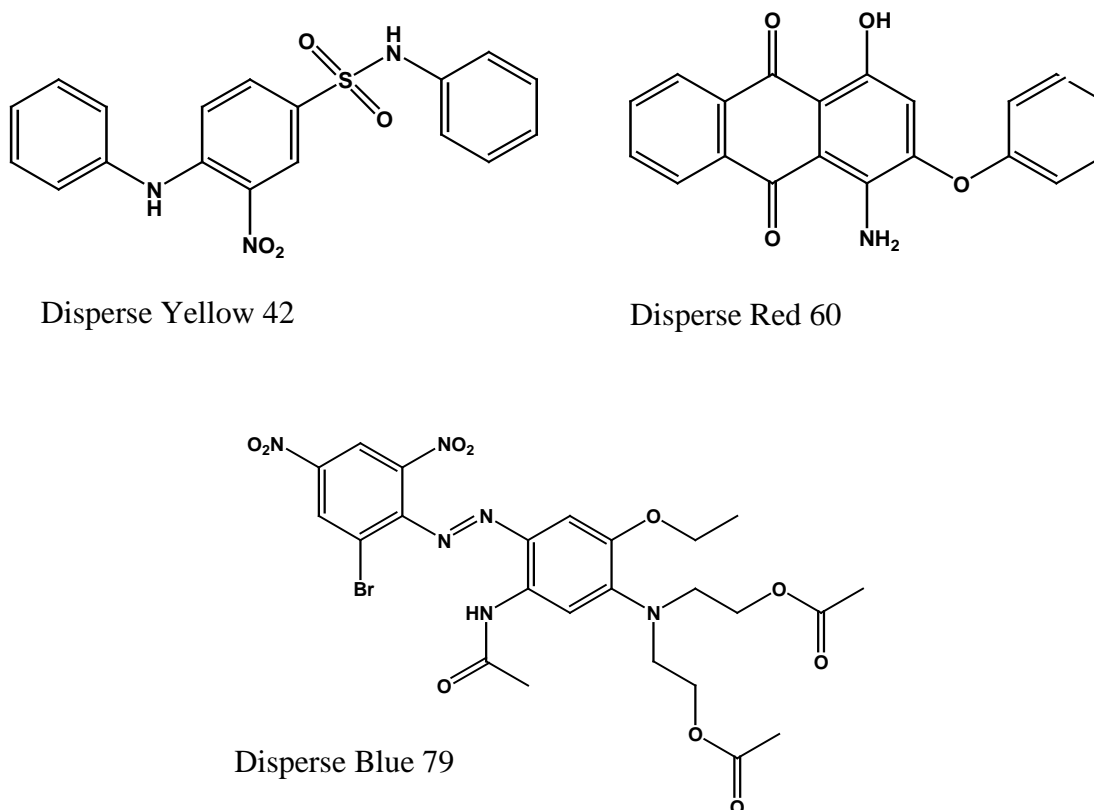


**Fig. 4.** Examples of reactive dyes commonly used for coloration of cellulosic textile materials

The dyes contain reactive groups, *e.g.*, monochloro-triazine (MCT), dichlorotriazine (DCT), or sulfatoethylsulfone (SES), that are reacted either by nucleophilic aromatic substitution or addition mechanisms. The process conditions (temperature and pH) are controlled carefully to minimize the amount of hydrolyzed dye that is formed by reaction with hydroxide. Hydrolysis generates hydroxytriazine and hydroxyethylsulfone functionalities from chlorotriazine and SES dyes, respectively. Because the dyes are very water-soluble, large amounts of salt are necessary to promote exhaustion. This salt is the major contaminant in the waste water from a plant that applies reactive dyes. After dyeing, the hydrolyzed dye must be removed by thorough scouring with a good surfactant to minimize problems with colorfastness to washing. Reactive Red 120 is an example of an azo dye containing two monochlorotriazine reactive groups, and Reactive Black 5 is an example of an azo dye with two sulfatoethylsulfone groups that will produce vinyl sulfones that are reacted with Cello<sup>®</sup> during fixation (Fig. 4).

*Disperse dyes and their use with polyester and cellulose ester fibers*

Disperse dyes were originally developed to color cellulose acetate and cellulose triacetate (Schuler 1980c). When polyester fibers were developed, these small, neutral nonionic dyes were successfully used to dye those fibers.

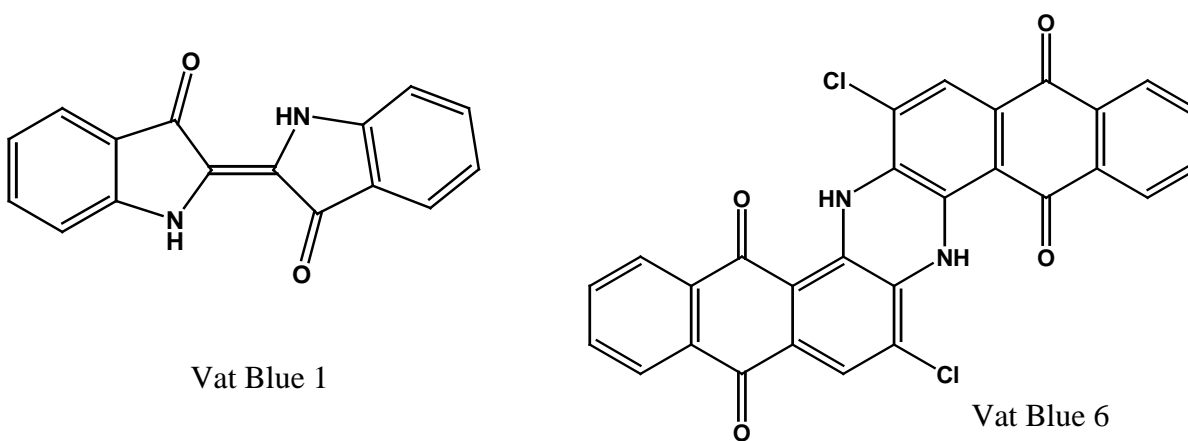


**Fig. 5.** Structures of disperse dyes commonly used to color cellulose ester-based materials

The bulk of disperse dyes contain one or more azo chromophores. Anthraquinone structures constitute the second most common chromophore. Disperse Yellow 42, a very common disperse dye used in automotive polyester, is an example of a nitrophenylamine chromophore. Dispersing agents, *e.g.*, lignin sulfonate and naphthalene sulfonic acid, are typically used in milling the disperse dye particles and keeping them dispersed during the dyeing process. In continuous dyeing, where the fabric is dried before heating (thermosolling) to swell the fiber and sublime the dye, an antimigrant is typically added to the dyebath. Effluent from batch disperse dyeings is likely to contain some unexhausted disperse dye, anionic dispersing agent, and some acetic acid. Discharge from continuous operations will contain disperse dyes, anionic dispersing agents, and antimigrants that are washed off the fabric after dyeing. Disperse Yellow 42, Disperse Red 60, and Disperse Blue 79 are examples of nitrophenylamine, anthraquinoid, and azo disperse dyes, respectively (Fig. 5).

#### *Vat dyes and their use with cellulosic fabrics*

Vat dyes are stored in an insoluble form and must be reduced to a soluble leuco form with sodium hydroxide and sodium dithionite (hydrosulfite) to be applied to cellulosic fabrics, either by exhaustion or in a continuous process. Once the leuco form has been applied to the fabric or yarn, it is oxidized back to the insoluble pigment form with hydrogen peroxide or, in the case of indigo, oxygen in the air. Indigo is unique, because it is applied to ropes or sheets of yarns in a dedicated indigo range. The yarns are dipped into boxes of reduced indigo and then exposed to air (skyed) to build up layers of the dye on the surface of the yarns. Often, the first dye box in a range may contain reduced Sulfur Black 1 (see next subsection) to cover some of the white warp yarns so that less indigo has to be used to achieve the desired denim shade. After oxidation, the dyed fabrics are soaped to aggregate the vat dyes and remove excess dye from the surface. Because indigo is on the surface of the yarns, it can be partially removed to give a desired “washed-down or worn” appearance. Indigo dyed yarns are not given an aggressive soaping. Indigo (Vat Blue 1) and Vat Blue 6 are examples of indigoid and anthraquinoid vat dyes (Fig. 6).



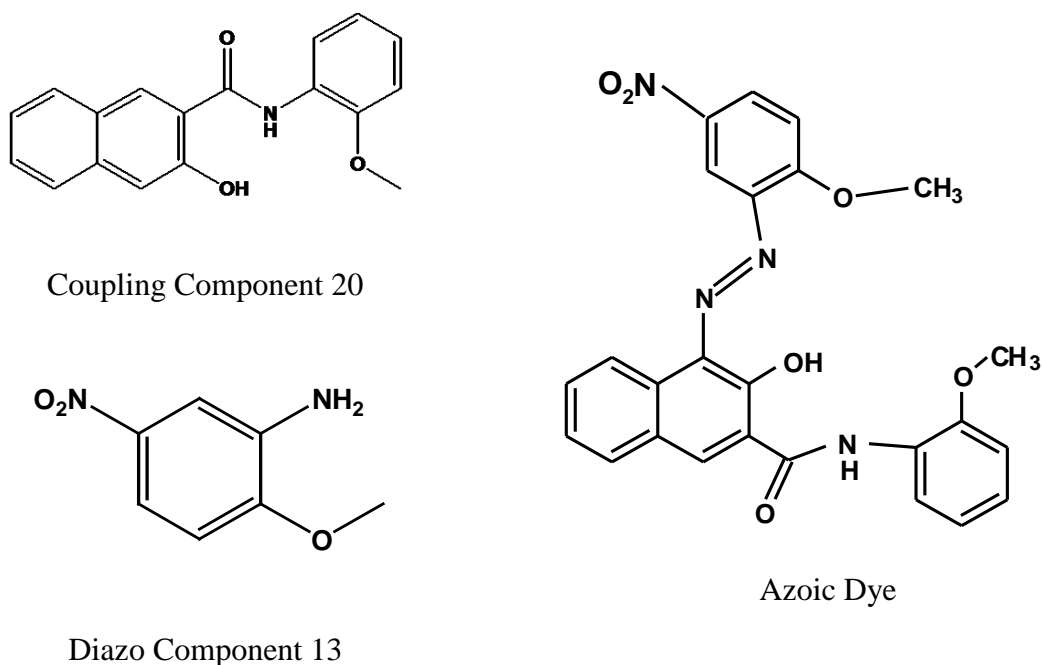
**Fig. 6.** Structures of representative vat dyes commonly used to color jeans and cotton ropes

*Sulfur dyes and their use with cellulosic fibers*

Sulfur dyes are similar to vat dyes with respect to their water insolubility and need to be reduced to a leuco form for application. Unlike the other classes of dyes, sulfur dyes typically are not represented by a single structure, but are a mixture of oxidized forms containing disulfide linkages. Reducing agents for sulfur dyes are typically sodium sulfide and sodium hydroxide or a sugar (Sandozol RDT, Clariant website) plus sodium hydroxide. Oxidation is effected with hydrogen peroxide, sodium bromate, or oxygen in air (sulfur on the bottom during indigo dyeing).

*Naphthol dyes and their use for cellulosic fibers*

Naphthol dyes are colorants that are formed inside the cellulosic fiber by a diazonium coupling reaction. Typically, a naphthol coupling component is solubilized in hot alkali, padded onto the fabric, and dried. The naphtholated fabric is passed through a pad bath containing a diazonium salt that has been formed by reacting a primary aromatic amine with nitrous acid. On contact with the naphthol, the diazonium salt undergoes an electrophilic aromatic substitution with the naphthol to form the naphthol (azoic) dye. Effluent from a naphthol dyeing operation will contain naphthols, aromatic amines, sodium nitrite, alkali, and acids. The azoic dye shown in Fig. 7 would be formed by the reaction between Coupling Component 20 and the diazonium salt from Diazo Component 13.



**Fig. 7.** Representation of a naphthol dye system used for coloration of cellulosic fibers. The moieties shown in the first two images (the coupling component and the diazo component) together comprise the azoic dye molecule (shown at right).

## SUBSTRATE CHARACTERISTICS AFFECTING DYE UPTAKE

Diverse cellulose-based materials have been found to be effective for the removal of dyes from aqueous solution, and factors affecting such processes will be reviewed in this section. To place the topic into context, the successful adsorption of a dye onto the selected substrate should be thought of as one step in an integrated program for water treatment. Other steps in bioremediation of dyes, to be considered near the end of this article, can include preparation and transportation of the biosorbent, bringing together the biosorbent and the polluted water, recovering the damp biomass, optional regeneration of the biosorbent after its use, and possible end-fates of the biosorbent. In addition, it is likely that future successful large-scale implementations involving biosorption of dyes will involve combinations of treatment stages, incorporating various more conventional approaches, such as those reviewed in the first section of this article. Given the fact that industrial implementation of biosorption for pollution control has been rather limited (Gadd 2009), it is important to carefully scrutinize factors that may contribute to its success in the future.

Considerable progress has been made by the authors of earlier review articles in dealing with the relationships between biosorbent characteristics and their ability to sorb different types of dyes (Sharma *et al.* 2011a). Classes of sorbent considered in these works have included the biomass of living and dead micro-organisms (Pearce *et al.* 2003; Akzu 2005; Vijayaraghavan and Yun 2008b), agricultural wastes (Hashem *et al.* 2007; Rafatullah *et al.* 2010), activated carbons prepared from agricultural waste biomass (Demirbas 2009), composites with chitosan (Ngah *et al.* 2011), and biosorbents in general (Srinivasan and Viraraghavan 2010). In addition Sun and Berg (2003) have reviewed the closely related field of using dyes for characterization of the charged nature of solid substrates. Further useful information can be found in review articles dealing with the dyeing of cotton and lyocell, *etc.* (*e.g.* Peters and Ingamells 1973).

### Tabulation of Biosorption Data by Dye Type and Sorbent Type

Table A, which due to its length is placed in the appendix of this article, represents an effort to bring together key data from published sources. Because there will be frequent references to Table A throughout this article, a brief description of its organization will be given here. The columns of the table give the dye class, the Color Index number (if known), the biomass class and type, whether the biomass was dried or heated, any significant modification of the biomass, the pH of maximum adsorption, the temperature at which the highest sorption was observed, the type of sorption isotherm giving the best fit, the adsorption capacity (mg/g), the rate law giving the best fit to kinetic data for adsorption, any functional groups found to be associated with sorption, whether adsorption was exothermic or endothermic, a column for any additional “key findings,” and a final column with the author-year information. Vertically, the items in the table are grouped together by dye class, starting with basic (Ba, positively charged) dyes, and then continuing through acidic (Ac), direct (Dr), reactive (Rx), vat (Vt), sulfur (Sr), disperse (Ds), and “not known” (Nk). If a numerical value appears in parentheses, that means that it was used as a default value, so it does not necessarily represent an

optimum condition for biosorption. A key to codes used in the table appears at the bottom of the table.

From Table A it is clear, first of all, that a great many different biomass-derived materials have been found to be effective for removal of dyes from aqueous solution. The table shows, for instance, many examples in which one type of cellulose-based materials is more effective than another with respect to its capacity to take up a specific type of dye. Given the great many ways in which biosorbent materials can differ from one another, especially if they have been chemically treated, such differences in removal capability according to biomass type should not be surprising. The goal of the present section is to consider whether there are certain characteristics of biomass that can help narrow the choice of materials that can be expected to be more effective for a given application.

### Classes of Lignocellulosic Material

As in the case of a previous article in this series dealing with biosorption of metal ions (Hubbe *et al.* 2011), it can be useful to group types of biomass into categories. Such grouping can help to frame the question as to whether certain such classes are more effective, as a rule, than others. Some of the most often studied types of cellulose-derived materials, for the purpose of dye removal, are activated carbon, fungal biomass, agricultural residues, wood (often in the form of sawdust), algae, aquatic plants, and textile fibers such as cotton and jute. Some important questions, when comparing classes of sorbent, can be listed as follows:

- Are there important differences between classes of cellulosic materials or derived products with respect to their capacity to adsorb different types of dyes?
- Are there large differences *within* given classes of such biosorbent materials?
- Can such differences in sorption capacity, whether they be within or between classes of biosorbent, be accounted for in terms of measurable quantities, such as apparent surface area, the density of charged chemical groups at the surfaces, or the relative hydrophobicity of the sorbent, *etc.*?

Tables 3A through 3D compare the mean, standard deviation, lowest reported value, and highest reported value of sorption capacity of samples from different classes of biomass for selected types of dyestuff, based on the present survey of literature. Table 3A shows data corresponding to adsorption of basic dyes, *i.e.* dyes that have a positive ionic charge in solution.

Before considering the mean values shown in Table 3A, it is important to consider the relatively large size of the standard deviations. For instance in the case of “Fungal biomass” the standard deviation was larger than the mean for the sorption capacity of basic dye onto different kinds of biomass. As will be discussed in more detail in subsections to follow, the wide ranges of the reported data can be tentatively attributed to differences in individual dyestuffs, details of preparation of the sorbents, and detailed conditions of testing. In any case it can be concluded, based on the standard deviations in Table 3A, that the “Class of Biomass” and the category “Basic dyes” can account for only part of the observed differences in sorbed amounts.



**Table 3A.** Reported Adsorption Capacities of Biosorbant Classes for Basic Dyes

Class of Biomass	Mean (mg/g)	St. Dev. (mg/g)	Lowest (mg/g)	Highest (mg/g)	No. of Reports
Pe = Peat	500	283	300	700	2
Ma = Marine plants	409	887	5	3200	12
F = Fungal	269	603	3	2670	26
AC = Activated carbon	240	268	2	1400	83
Pl = Plants, non-ag	179	273	20	833	9
N = Nuts, pits, and shells	120	112	15	360	10
Ag = Agricultural residue	106	112	3	556	52
Al = Algae	104	109	9	417	16
FA = Fly ash	90	99	3	181	4
B = Bacteria	71	57	8	190	12
W = Wood	70	87	1	425	33
Sl = Sludge	28	28	5	60	3
Tx = Textile materials	17	11	5	25	3
Ce = Cellulose	11	8	2	17	3

\* Values taken from Table 1 (appendix), generally using the highest value reported in a given study, *i.e.* the optimum conditions of adsorption

Because the data prepresented in the table come from different studies, each having its own goals and constraints, caution is advised when comparing mean values – especially when a mean value is based on just a couple of studies (see final column). To broadly summarize the results in Table 3A, relatively high amounts of basic dyes tended to be taken up, on average, by peat (based on just two reported values), marine plants, fungal biomass, and activated carbon samples. Much lower amounts were taken up, on average, by such sorbents as wood powder, sludge, fly ash, textile fibers (cotton and jute), and pure cellulose substrates.

As will be suggested in the next subsection, differences in accessible surface area can be expected to account for much of these differences in sorption capacity. For instance, the word “activated” in the term “activated carbon” implies a process in which the surface area of the material is greatly increased through development of fine-scale porosity. By contrast, the relatively thick and dense cell walls of wood fibers can be expected to limit how much of the material, per unit mass, can be approached by dye molecules to within molecular dimensions from the cellulosic material.

Table 3B shows similar results for acid dyes, which bear a negative charge in solution.

**Table 3B.** Reported Adsorption Capacities of Biosorbant Classes for Acid Dyes

Class of Biomass	Mean (mg/g)	St. Dev. (mg/g)	Lowest (mg/g)	Highest (mg/g)	No. of Reports
Al = Algae	514	577	71	1360	4
Pl = Plants, non-ag	481	757	47	2000	6
AC = Activated carbon	315	519	4	2700	30
Tx = Textile materials	293	306	13	800	5
F = Fungal	218	278	1	1111	24
W = Wood	166	152	19	412	5
Ag = Agricultural residue	153	294	1	1072	19
Ma = Marine plants	104	66	22	244	9
B = Bacteria	102	11	91	112	4
Sl = Sludge	40	14	30	50	2
N = Nuts, pits, and shells	25	24	5	55	4
FA = Fly ash	19		19	19	1
Ce = Cellulose	12	10	3	25	4

A particularly interesting aspect of the results in Table 3B is the extensive re-ordering of the rows relative to Table 1. In other words, different classes of biomass tended to be most effective for sorption of acid dyes, as compared to basic dyes. While some of these apparent differences might be due to chance variations in the details of different studies, such results also may shed light on the mechanisms (to be discussed in greater detail in later sections).

Since acid dyes in solution of near-neutral pH have the same sign of charge as the surfaces of most unmodified biomass, one can expect that dye adsorption will be highly dependent on either (a) various modifications of the substrate, including cationization, (b) bonding mechanisms other than attraction of opposite charges, and (c) the fact that most of the reported studies showed optimum biosorption of acid dyes in a pH range between 1 and 2.

However, before one gets too comfortable with the statements just made, let us consider the results for another class of dyestuffs that generally have a negative charge in solution, the reactive dyes. Table 3C shows those results, again abstracted from Table A in the appendix. In some respects the results in Table 3C are more similar to those in Table 3A, despite the difference in sign of charge of the dyes in question. Recently Tigini *et al.* (2012) were able to demonstrate a rough correlation between the chemical composition of natural fungal biomass types and their ability to take up dyestuffs of a specific sign of charge. Those samples that were rich in amino groups were found to be more effective for removal of negatively charge dyes, such as reactive dyes. Another way to compare the sorption capacities of different biomass classes is to include all of the dye classes together. Thus, the data prepresented in Table 3D includes not only all of

the data in the previous three tables, but also reported results for the following types of dyestuffs: direct, vat, sulfur, and disperse.

**Table 3C.** Reported Adsorption Capacities of Biosorbant Classes for Reactive Dyes

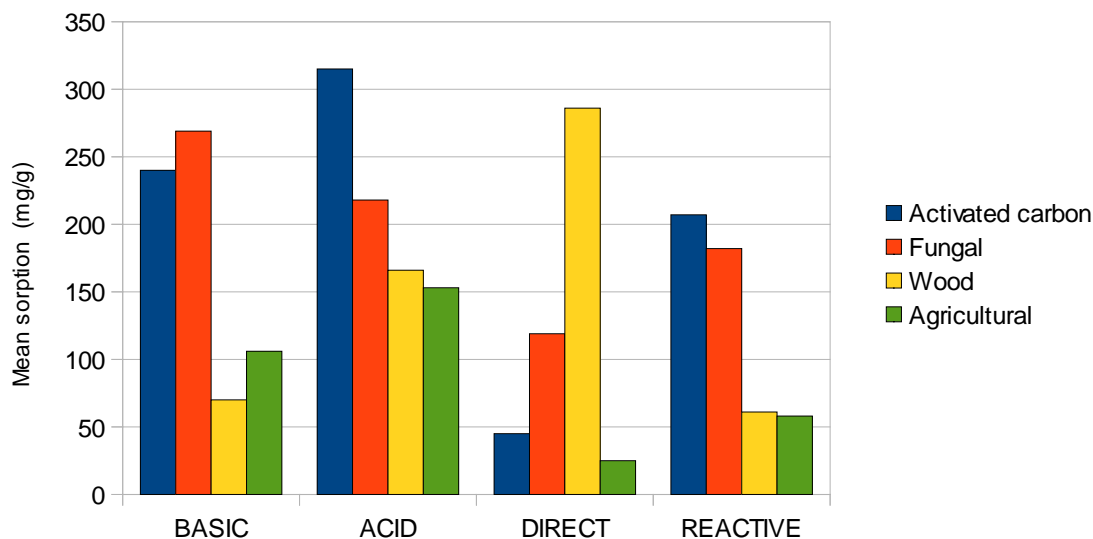
Class of Biomass	Mean (mg/g)	St. Dev. (mg/g)	Lowest (mg/g)	Highest (mg/g)	No. of Reports
AC = Activated carbon	207	211	3	714	15
Al = Algae	199	151	44	420	6
F = Fungal	182	200	4	1008	35
B = Bacteria	171	123	8	485	25
Sl = Sludge	71	50	7	127	7
W = Wood	61	88	4	249	8
Ag = Agricultural residue	58	96	2	320	18
FA = Fly ash	43	55	4	135	7
Ce = Cellulose	42 -		42	42	1
N = Nuts, pits, and shells	38 -		38	38	1
Ma = Marine plants	30	26	5	67	7

**Table 3D.** Reported Adsorption Capacities of Biosorbant Classes for All the Reported Data

Class of Biomass	Mean (mg/g)	St. Dev. (mg/g)	Lowest (mg/g)	Highest (mg/g)	No. of Reports
Pe = Peat	500	283	300	700	2
Pl = Plants, non-ag	300	520	20	2000	15
AC = Activated carbon	247	331	2	2700	136
F = Fungal	211	374	1	2670	90
Ma = Marine plants	203	564	3	3200	31
Al = Algae	182	268	6	1360	27
W = Wood	152	426	1	2998	51
B = Bacteria	135	110	8	485	41
Tx = Textile materials	132	221	5	800	13
Ag = Agricultural residue	98	160	0	1072	99
N = Nuts, pits, and shells	81	97	5	360	17
FA = Fly ash	74	110	3	400	15
Sl = Sludge	55	44	5	127	12
Ce = Cellulose	14	12	2	42	15

As a point of reference, the overall mean sorption capacity, representing 564 reported values (from Table A in the appendix), was 168 with a standard deviation of 256.

Another way to address the question, “did the class of dye generally make a difference” is to plot mean sorption amount versus dye class. Figure 8 shows such a plot, focusing on four of the most widely studied classes of biosorbent. Again, marked differences are apparent, and at least part of them are unlikely to be explainable in terms of the general classes of biosorbent and or dye. It is important to note, however, that efficient uptake of dyes was achieved for each class of dyestuff – though not equally on every class of biosorbent. The relatively high capacity of the “wood” material for direct dyes has a special explanation: some of the tests represented in that mean value corresponded to cellulose substrates that had been derivatized to give them a positive surface charge. Another factor that could be expected to bias the data is the fact that acid dyes are known to be more highly soluble and inherently difficult to retain; thus, researchers attempting to adsorb acid dyes would be more likely to take more extreme measures (substrate derivatization, use of very low pH values, *etc.*) in an attempt to remove the dyes from solution.



**Fig. 8.** Mean sorbed amounts of four classes of dyestuff onto four of the most commonly studied classes of cellulosic-derived sorbents

### Surface Area

It is commonly assumed that the amount of dye taken up by a substrate ought to be proportional to the surface area. This assumption underlies a number of procedures in which dye adsorption has been used as a means of determining the surface areas of various solids (Kaewprasit *et al.* 1998; Kim *et al.* 2004; Ibbett *et al.* 2006a). In the case of cellulose-based materials, however, one needs to carefully consider the definition of “surface”. For instance, the external surface area of wetted kraft fibers has been found to account for only a minor fraction of adsorbed cationic polyelectrolytes of sufficiently low

molecular mass (Wågberg and Ödberg 1989). Various studies have shown that, given time, even relatively large molecules are able to permeate deep within the cell walls of wood fibers (Horvath *et al.* 2008; Wu *et al.* 2009).

The superior adsorption capacity of certain activated carbon and related products has been attributed to their relatively high surface area per unit mass (El-Sharkawy 2001; Huang *et al.* 2001; Nasr *et al.* 2006; Asadullah *et al.* 2010; Ling *et al.* 2011). For instance, Meng *et al.* (2008) attributed superior uptake of dyes by a certain activated carbon sample to the presence of a large surface area associated with micro-pores in the range of 1 to 1.5 nm.

An early study of the relationship between surface area and dye uptake was undertaken by Thode *et al.* (1953). Their results showed a positive correlation between pulp refining and dye uptake. Because refining of pulp is known to delaminate the fibers both at their outer surfaces and internally, one possible explanation for the results is that dye adsorption increased in direct proportion to the accessible surface area. But it is also possible to envision a more complex mechanism whereby the delamination caused by refining helps the dye to diffuse more rapidly or to a greater extent within the bulk of the water-swollen cell wall.

Work by Ladchumananandasivam *et al.* (1994) suggested that dyes would adsorb onto the surfaces of crystalline nano-domains in cotton. In addition, it would be reasonable to suspect that dyes, depending on their molecular size and solubility characteristics, may permeate into the amorphous regions of cellulose (Kreze *et al.* 2002). Timofei *et al.* (1994) in a simulation study of binding sites, concluded that both crystalline and amorphous cellulose sites are likely to bind with dye molecules. Saravanan *et al.* (2009) in their review of the biopolishing of cellulosic fibers with cellulases, cited various literature suggesting that relatively large dye molecules adsorb mainly on the outer, fibrillated parts of cellulosic fibers, which are essentially the same areas that tend to be cleaned from the fiber surfaces during the enzymatic pretreatments. However, depending on the conditions and length of pretreatment, enzymatic action appeared able to open up fresh surfaces, thus increasing dye uptake in some cases.

If one assumes, at least as an approximation, that dye sorption is proportional to the external surface area of cellulosic particles, then one would expect to see the following kind of relationship,

$$\Gamma = k_1 \bullet (\text{specific area}) = k_2 / d \quad (1)$$

where gamma represents the amount adsorbed per unit mass of sorbent,  $k_1$  and  $k_2$  are constants, and  $d$  represents the diameter of a particle of the biosorbent. Consistent with this relationship, a number of studies have shown an inverse relationship between substrate particle size and the amount of dye removed per a given mass (Poots *et al.* 1976; Ibrahim *et al.* 1997; Annandurai and Sheeja 1998; Ho and McKay 1999; Robinson *et al.* 2002b,c; Banat *et al.* 2003; Ho *et al.* 2005a; Preethi *et al.* 2006; Laasri *et al.* 2007; Dizge *et al.* 2008; Dogan *et al.* 2008, 2009; Gercel 2008; Ponnusami *et al.* 2008; Ofomaja 2008, 2009; Wang *et al.* 2008b; Gad and El-Sayed 2009; Gupta *et al.* 2010; Laohaprapanon *et al.* 2010; Li *et al.* 2010; Ouazene and Sahmoune 2010; Nethaji and Sivasamy 2011). Though none of the listed studies attempted to establish the validity of

the relationship shown in Eq. 2, most data suggest a much weaker dependency on particle size. For example, Nethaji and Sivasamy (2010; 2011) reported roughly a 50% increase in dye sorption per unit mass of activated carbon associated with a ten-fold decrease in particle size from 1 mm to 0.1 mm. Such deviations from Eq. 2 suggest an important role of internal pore spaces in the adsorption of dyes, depending on the nature of the sorbent.

### Pore Size Distribution

Ibbett *et al.* (2007a) introduced the concept of “accessible volume” to account for dye uptake by cellulosic fibers having different histories of drying. Never-dried fibers appear to have spaces between cellulose crystallites where dye molecules are able to permeate. However, such spaces are known to close up in a semi-permanent manner when cellulosic fibers are dried (Stone and Scallan 1966; Hubbe *et al.* 2007). Thus Ibbett *et al.* (2007a) observed a reduction in swellability and dye uptake associated with drying of lyocell fibers. Likewise, some authors (Yang *et al.* 1995; Ibbett *et al.* 2007b) found that dye uptake was reduced by incorporation of crosslinking in the fibers, a measure that reduced swelling in water. Chen *et al.* (2005) observed similar effects; however their results were found to depend on the nature of the crosslinking. The longer lengths of cross-linking elements in so-called “steep-pad-drycure” cotton fabrics allowed higher dye uptake in comparison to a more standard treatment for textiles used in manufacture of never-iron clothing.

Various authors have shown that both the swelling and dye uptake capacity of previously dried cellulosic fibers can be restored by alkaline treatment. For instance, Ibbett *et al.* (2007b) found that it was possible to restore dye uptake of crosslinked cotton to a high level by caustic treatment, which was able to overcome the cross-linking and reswell the fibers. Siroky *et al.* (2011) likewise observed greater dye uptake in fibers that had been swollen by treatment with NaOH. Presumably such swelling can be considered as being equivalent to the opening up of fine pores suitable for dye adsorption within the cell walls.

### Ion Exchange Capacity

Though analyses based on surface area generally assume that there is a one-to-one correspondence between surface area and dye uptake, such studies usually provide little evidence of whether the available surfaces are completely covered by dye molecules, and even if so, what kinds of forces are responsible for the adsorption. As will be shown, there is considerable evidence in support of an electrostatic mechanism of binding, especially in cases involving charged dye species. In other words, charged sites at the wetted surfaces of cellulosic materials appear to bind oppositely charged dye molecules by an ion exchange mechanism (Mathews *et al.* 2004; Suteu *et al.* 2010; Tan 2010). Such a mechanism can explain, for instance, why the uptake of basic dyes can be increased greatly by derivatizing cellulosic materials to increase the amount of carboxylic groups on their surfaces (El-Hilw 1999; Abo-shosha *et al.* 2002). Likewise, the uptake of anionic dyes can be greatly increased by cationizing the cellulosic substrate (Waly *et al.* 1998; Baouab *et al.* 2000, 2001; Hashem and El-Shshtawy 2001; El Ghali *et al.* 2010). In a reverse sense, Vijaraghavan *et al.* (2008a) demonstrated the same mechanism by showing higher sorption of negatively charged dye onto fermentation waste biomass after

the latter had been decarboxylated. Ofomaja (2009) argued that an ion exchange mechanism helps to explain the magnitudes of binding energies between dyes and surface sites.

Faria *et al.* (2004) carried out a study in which an activated carbon sample was post-treated in order to alter the balance between acidic and basic sites on its surface. As would be expected based on electrostatic attractions, the carbon samples that were rich in acidic sites tended to be more effective for basic dyes. On the other hand, carbon that was rich in basic sites had higher affinity for anionic dyes. However, the trends were not clear cut, so the cited authors concluded that other factors, such as dispersion forces, must play a major role in adsorption of dyes in such systems.

### Zeta Potential

Another way to demonstrate the role of ionic charges of the substrate for the adsorption of ionic forms of dye molecules is by measuring the zeta potential. This quantity can be defined as the electrical potential at a hydrodynamic slip plane adjacent to the substrate surface (Hiemenz and Rajagopalan 1997; Sun and Berg 2003; Kumar and Teli 2007). Strong relationships have been found between zeta potential and the uptake of various dyes from solution (Yenikaya *et al.* 2009; Tian *et al.* 2010). In other cases zeta potential has been used to characterize the charge modification of surfaces as a means of increasing the sorption of oppositely charged dyes (Petzold *et al.* 1997; Kumar and Teli 2007; Akar and Divriklioglu 2010). As further confirmation, yet other researchers have noted changes in zeta potential resulting from dye adsorption onto cellulosic substrates (Espinosa Jimenez and Gonzalezcaballero 1991; Stana *et al.* 1995).

A further way to demonstrate the influence of the substrate's zeta potential on dye sorption is by considering pH relationships. Entries in Table A show examples in which basic dyes, which have a positive charge in solution, are preferentially adsorbed at relatively high pH; under such conditions the carboxylic acid groups at the surface of various cellulose-related surfaces would be dissociated, yielding a negative net charge. By contrast, adsorption of negatively charged dyes, such as direct dyes and most reactive dyes, has been found to be maximized at very low pH in many cases.

### Hydrophobic Aspects of Cellulose

Given the substantially hydrophobic character of the chromophoric groups in typical dyestuffs, it is reasonable to expect there to be a relationship between sorption and the hydrophobicity of the substrate. The performance of typical dye molecules in various applications depends on a delicate balance between hydrophobic and hydrophilic characteristics. On the one hand, the colorant must be sufficiently solubilized so that it can be distributed during the application. On the other hand, it needs to have a sufficiently strong tendency to come out of solution or suspension and adsorb onto the material that needs to be colored. The structure of typical dye materials is consistent with a strong tendency to be at least partly hydrophobic; extended conjugation (alternating double and single carbon-carbon bonds, often including aromatic groups) makes it possible for an organic molecule to absorb visible light. Such molecular structures contribute toward the hydrophobic character of a molecule. Most dye applications call for dissolving of the material as an aqueous solution. Solubility can be achieved by

incorporating a suitable number of charged groups such as sulfonates or amines into the dye molecule. If there are too few such groups, then the dye will be difficult to dissolve and thereby distribute to the target material. If there are too many such groups, then the dye may tend to be too soluble, preferring to remain in solution rather than adsorb onto the substrate.

Even though the cellulose molecule has three hydrophilic –OH groups per pyranose unit, it is not necessarily considered to be highly hydrophilic (Biermann *et al.* 2001). The somewhat hydrophobic nature of cellulosic materials can be at last partly attributed to participation of most of the –OH groups in semi-permanent intra-molecular and inter-molecular hydrogen bonding (Mann and Marrinan 1958; Kadla and Gilbert 2000). However, recent research suggests that cellulose crystallites can present sharply contrasting faces; one such face can account for the fact that regenerated cellulose film allows water droplets to spread to a lower contact angle than films of starch or polyvinyl alcohol, whereas the other face can account for the ability of cellulose to adsorb various oils effectively (Yamane *et al.* 2006). On the other hand, the presence of hemicellulose in many lignocellulosic materials contributes to making the material more hydrophilic. This is shown by the fact that wood takes up less water after hemicellulose has been removed by extraction (Zhang *et al.* 2011).

### Hydrophobic Aspects of Lignin

Due to lignin's relatively hydrophobic character and its content of many aromatic groups, there is reason to expect typical dye molecules to have good affinity with this component of plant material. The ability of isolated lignin materials to sorb dyes has been well demonstrated (Nikiforov 1985; Wardas and Lebek 1994; Lebek and Wardas 1996; Liu *et al.* 2005, 2006; Suteu *et al.* 2010). For instance, Lebek and Wardas (1996) showed that lignin left over as a byproduct of vanillin production has a good affinity not only for cationic dyes, but also for reactive dyes, which are anionic in character. The cited authors attributed a higher adsorption tendency of cationic dyes to interaction with the negatively charged sites on the lignin. Zafar *et al.* (2008) used evidence from infrared adsorption spectroscopy to assert that the adsorption of methylene blue was associated with interaction with the C-O groups and aromatic groups within lignin.

A strong positive correlation was observed between adsorption of basic dye (either methylene blue or crystal violet) and the lignin content of spruce thermo-mechanical pulp (TMP) (Peterlin *et al.* 2009b). However, a negative correlation was found between methylene blue dye sorbed amount and the lignin content of spruce mechanical pulp that had been subjected to different levels of oxidation (Peterline *et al.* 2009a). Such behavior is consistent with the initial increase in content of carboxylate groups, resulting from oxidation. One should keep in mind, however, that chemical pulping and bleaching processes can be expected to remove products of oxidation from fiber surfaces. As a result, low-yield pulp fibers, from which lignin, extractives, and some of the hemicelluloses have been removed, can be expected to have less affinity towards basic dyes.

In the case of toluidine blue dye, van de Ven *et al.* (2007) were able to distinguish between two factors that promoted its adsorption. On the one hand, the cationic nature of the dye favored its sorption onto the anionic groups of wood pulp fibers. On the other



hand, pulps that contained lignin above a critical level were able to adsorb more of the cationic dye, above the level that would be predicted by charge stoichiometry. The latter observation supports a major role being played by dispersion forces and the hydrophobic groups on both the dye molecules and the substrate surfaces.

### Modifications to the Cellulosic Substrate

A great many published studies have considered whether it is advantageous to modify cellulose-derived substrates in order to achieve greater removal of dyes from aqueous systems. Leaving aside the question of whether there might be other reasons for such modification (such as better storage stability, easier handling, *etc.*), it would make sense to place highest priority on the use of inexpensive resources that are already highly effective for dyes adsorption and do not require any further treatment. Therefore, the first treatments to be considered below will include “non-treatment” and relatively simple treatments such as drying.

### Whether the Substrate is Alive

Various studies have shown, perhaps surprisingly, that dye uptake is often increased by treating biomass in various ways so that it is no longer living. Table A indicates many cases in which either dry heating or autoclaving increased the adsorption capacity of the substrate for the target dye. The moist heating within an autoclave increased dye sorption, or at least still gave favorable results, according to several sources (Binupriya *et al.* 2007, 2008b; Patel and Suresh 2008; Prigione *et al.* 2008; Seyis and Subasioglu 2008; Sadhasivam *et al.* 2009). Prigione *et al.* (2008) noted that in addition to taking up more dye, the inactivated biomass did not require a continuous supply of nutrients, it was not sensitive to the toxicity of dyes, it tended to be stable during storage, and it could be regenerated and reused many times. Kaushik and Malik (2010, 2011) showed that the ability of fungal biomass to take up dye was highly dependent on the nutrients employed during production of the sorbent material; however their work did not clearly show whether or not continued viability of the substrate was an advantage.

Although the present review article is mainly concerned with adsorption as a means of removing dyes from aqueous solution, there has been a substantial body of published studies dealing with effects of biological processes on the chemical breakdown of dyes. Some key review articles dealing with such studies are available (Banat *et al.* 1996; Rodríguez-Couto 2009; Fu and Viraraghavan 2001a). In particular, many enzymes, such as laccase varieties, have been shown to be highly effective in breaking down dye materials to colorless forms (Walker and Weatherley 2000; McMullan *et al.* 2001; Acuner and Dilek 2004; Rao and Venkateswarlu 2006; Madhavi and Lele 2009; Lamia and Neji 2010; Boran and Yesilada 2011; Galai *et al.* 2012; Guisado *et al.* 2012; Kunjadia *et al.* 2012). Though many enzymes can persist even after the producing organism is no longer viable, only a living organism can produce new enzymes. On the other hand, even in a case where the enzymes from living bacteria were actively breaking down dye molecules, up to 19% of the decoloration still could be attributed to biosorption (Walker and Weatherley 2000). El-Rahim (2006), whose study considered both living fungal decolorants and low-cost cellulosic biomass concluded that the latter approach – relying on biosorption – holds considerable promise.

## Drying

As can be seen from Table A, the great majority of the cited studies employed drying, usually in the presence of heat, as the first step in the preparation of the substrate. There are practical reasons for this. On the one hand, it is reasonable for researchers to attempt to establish a reproducible starting point. Because the detailed history of moisture content of a plant-based material is often unknown, a standardized procedure of drying will presumably allow later researches to repeat the work. Furthermore, a dried material is likely to be more suitable for long-term storage, being less subject to decay. In addition, there may be savings in transportation, since the mass will be reduced by drying. The downside of this procedure is that technologists may be inadvertently sacrificing a portion of the adsorptive capacity of the substrate. As mentioned earlier, when discussing surface area and accessible pores in cellulosic substrates, there is a tendency for many mesopores within cellulosic materials to close up in a hard-to-reverse manner during drying (Stone and Scallan 1966). Thus it would be reasonable to expect such treatments to have a significant effect on dye uptake.

Only a few studies have directly compared dried *vs.* never-dried substrates with respect to dye uptake. In the case of lyocell fibers, reductions in swellability and dye uptake were observed as a consequence of drying (Ibbett *et al.* 2007a). Inglesby and Zeronian (1996) reported similar observations for mercerized cotton. Given the importance of this issue, one can hope that significant future work will be undertaken in this area.

## Immobilization

Another practical way to improve the handling of a finely divided biosorbant material is to immobilize it onto fibers or other suitably permeable substrate (Rodríguez Couto 2009). It is typical for bacterial and fungal cells to become at least partly immobilized by being cultured in the presence of a suitable substrate. By contrast, few if any researchers have considered the possible use of chemical flocculants to immobilize fine particles or microbial cells onto a cellulosic support for purposes of adsorbing dyes from effluent. Such approaches may be able to minimize problems associated with separation of very fine solids from an aqueous solution after use. Good performance of immobilized biomass products in the sorption of various dye products has been observed (Pazarlioglu *et al.* 2005; Ramsay *et al.* 2005; Iqbal and Saeed 2007; Wang and Hu 2007; Ertugrul *et al.* 2008; Tan *et al.* 2008b; Vijayaraghavan and Yun 2008a; Vijayaraghavan *et al.* 2008a-c; Chen *et al.* 2009; Chu *et al.* 2009; Maurya and Mittal 2009; Saeed *et al.* 2009; Asgher and Bhatti 2010; Mao *et al.* 2010). However, many researchers have observed a partial loss of sorption capacity, *i.e.* the amount of dye removal was reduced in comparison with the freely suspended sorbent (Vijayaraghavan *et al.* 2008b; Vijayaraghavan and Yun 2008d; Asgher and Bhatti 2010; Binupriya *et al.* 2010). Binupriya *et al.* (2010) attributed the poor sorptive performance of immobilized bacterial cells to poor mass transfer characteristics of the polymer matrix that they used. Likewise, Vijayaraghavan and Yun (2007a) and Mao *et al.* (2010) observed slower sorption onto immobilized citric-acid-treated bacterial material, compared to the freely suspended sorbent. By contrast, Saeed *et al.* (2009) reported a case in which immobilization had a favorable effect on dye sorption. Since the matrix employed for immobilization of the

fungal mold biomass in that case was loofa sponge, which itself is a high-surface-area cellulosic material, it is understandable that the latter material may have been contributing a significant amount to the uptake of dye. Wang and Hu (2007) compared several different kinds of support, and they found the best results when the surface texture of the immobilized bed was loose and finely porous.

### Derivatization

Some examples of chemical treatments were already mentioned when discussing the importance of the charged nature of the cellulose-based substrate in the sorption of various ionically charged dye species. Many examples are shown in Table A. Derivatization can be especially advantageous for sorption of anionic dyes (*e.g.* acidic, direct, reactive), which can be inherently challenging to collect on the negatively charged surfaces of unmodified lignocellulosic materials (Waly *et al.* 1998; El-Hilw 1999; Shi *et al.* 1999; Low *et al.* 2000; Hashem and El-Shishtawy 2001; Bouzaida and Rammah 2002; Lim *et al.* 2003; Hashem *et al.* 2006; Baouab *et al.* 2007; Temuz *et al.* 2007; El Ghali *et al.* 2010; Yigitoglu and Temocin 2010). The opposite approach of grafting additional acidic groups onto a biomass substrate was used by Won *et al.* (2009a), who observed increased removal of basic dyes. Fang *et al.* (2010) took a somewhat different approach by preparing a lignin-based cationic flocculant, which proved to be effective for the flocculation and removal of various anionic dyes from solution.

### Adsorption of Cationic Polyelectrolytes

As a possible alternative to chemical derivatization, researchers have considered the practical approach of treating the cellulosic sorbent with a solution of a cationic polyelectrolyte, which, due to its charge difference, can be retained efficiently on most biomass-derived materials. Authors have reported enhanced uptake of anionic dyes when using such an approach by the pretreatment of the substrate with chitosan, a cationic natural polyelectrolyte (Ngah *et al.* 2011). Kikhulnumchai *et al.* (2008) achieved the same effect in a more permanent manner by oxidation of cellulose fabric, followed by reductive amination in the presence of chitosan.

## SOLUTION CHARACTERISTICS AFFECTING DYE UPTAKE

Aqueous solution characteristics have been shown to have major influence on the uptake of dyes by various cellulose-based materials (Aksu 2005; Safa and Bhatti 2010). Factors to be covered in this section include pH, ionic strength, divalent ions, surfactants, and various fixatives added to the solution phase.

### Concentration of the Dye and Time of Contact

The initial concentration of dye in contaminated water provides an important driving force to overcome mass transfer resistance of molecules between the aqueous and solid phases (Dogan *et al.* 2006; Sharma *et al.* 2008). In physical adsorption most of the adsorbate species are adsorbed on the interface within a short interval of contact time.

However, a longer contact time, often on the order of a few hours, is often needed for the attainment of equilibrium.

Adsorption experiments have been conducted to study the effect of the initial dye concentrations of adsorbent in the solutions on the rate of dye adsorption onto adsorbent. Such experiments are usually carried out at a fixed adsorbent dose and at different initial dye concentrations of adsorbent for different time intervals and at fixed pH and agitation speed. The percentage removal generally will increase with a decrease in initial concentration. However, the dye uptake per unit weight of the adsorbent typically increases with the increase in the initial dye concentration (Hameed *et al.* 2008b,c). Similar observations have been reported by several workers for the adsorption of dyes and metal ions on bamboo, rubber sawdust, palm shell, and durian peels (Hameed *et al.* 2007; Adinata *et al.* 2007; Hameed and Hakimi 2011). An explanation for the latter trend is that increasing the initial dye concentration increases the number of collisions between the dye ions and the adsorbents (Weber and Morris 1963; Eckelfelder 1980). Bulut and Aydin (2006) studied methylene blue adsorption on wheat shells and reported around 135 minutes equilibrium adsorption time. However, the initial dye concentration often does not appear to have a significant effect on the equilibrium time, depending on the dyes and adsorbents used (Hameed *et al.* 2007, 2008b,c; Sharma *et al.* 2011b; Singh *et al.* 2011).

At higher initial concentration, the available adsorption sites of adsorbent become relatively fewer with the passage of time. Large numbers of surface sites are available for adsorption at initial stages, whereas and after a lapse of time, the remaining surface sites may be those that more difficult to occupy due to inaccessibility. Also, the remaining surface sites can become more difficult to occupy because of repulsion between the already-adsorbed solute molecules and those still in the bulk phase.

The initial concentration of adsorbates and contact time between the adsorbates and the adsorbents are of practical importance when studying removal of dyes from their aqueous solutions by adsorption on solid adsorbents. A given mass of the adsorbent, depending on its preparation, can adsorb only a fixed amount of solute. Thus, in case of a more concentrated solute, the volume of effluent that can be effectively treated is less. Alternatively, if the volume of effluent is kept the same, proportion of dye molecules that are adsorbed becomes low when treating concentrated dye solutions (MacKay and Ho 1999). The contact time required to attain equilibrium tends to be longer for premium, highly porous sorbent materials such as activated carbon; by contrast the equilibrium time is shorter in the case of non-porous adsorbates.

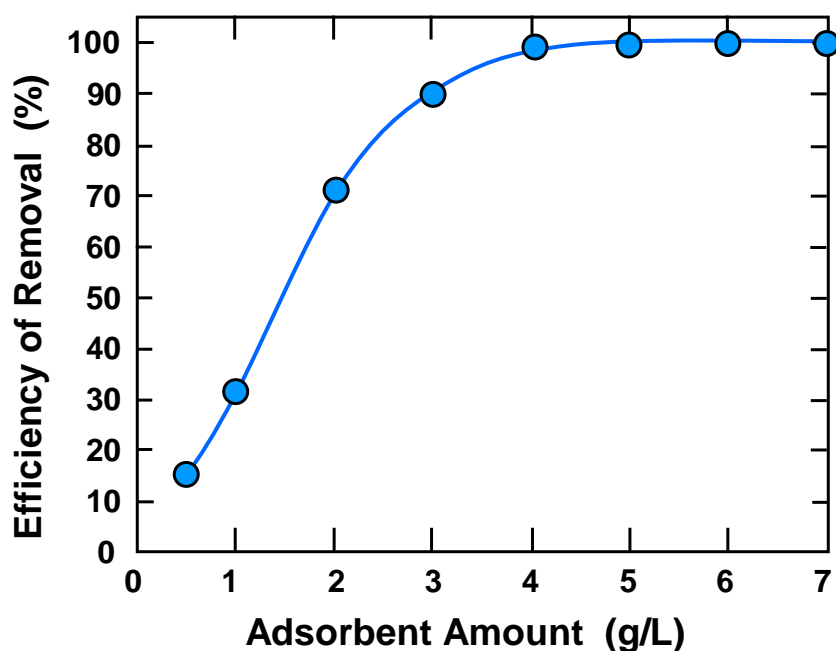
Some researchers have attributed differences in the time required for dye uptake to differences in the mechanism or energetics of sorption. Thus, it has been stated that in physical adsorption, most of the adsorbate species are adsorbed on the interface within a short interval of contact time (Kavitha and Namasivayam 2007; Hameed *et al.* 2008c; Khraisheh *et al.* 2002). By contrast, strong binding of adsorbate with adsorbent, which may involve migration of dye molecules to interior sites, can require a longer contact time for the attainment of equilibrium.

At lower dye concentrations, the ratio of the initial number of dye molecules to the available surface area is low, and subsequently the fractional adsorption becomes independent of the initial concentration. However at higher concentration the available sites of adsorption become fewer, and hence the percentage removal of adsorbent is

dependent upon initial concentration. This may be due to the fact that at lower concentrations almost all the dye molecules were adsorbed very quickly on the outer surface, but further increase in initial dye concentration leads to fast saturation of adsorbent surfaces, and thus most of the dye adsorption depends on a slower process of diffusion inside the pores.

### Effect of Adsorbent Amount

The amount of adsorbent used in comparison to the volume of contaminated water determines the capacity to absorb dyes. Higher dye removal at increased biosorbent dose may be attributed to availability of a large number of active sites for the same number of adsorbate molecules. Experimental confirmation of this principle has been shown, for instance, by Mall *et al.* (2005) for the removal of methylene blue and malachite green by adsorption on coconut coir activated carbon and rice husk activated carbon. Sharma *et al.* (2009) also reported similar behavior for the adsorption of methylene blue on coconut coir activated carbon. Figure 9 provides an example in which the effect of adsorbents dosages were studied while keeping all other experimental conditions constant and varying the initial dye concentration (Deng *et al.* 2009). The effect of adsorbent amount on percentage removal and uptake of dye is shown.



**Fig. 9.** Effect of dose of adsorbent on the removal of methylene blue (pH = 7.0,  $C_0$  = 1500 mg/L, contact time=2h); Data source (redrawn): Deng *et al.* (2009).

The results in Fig. 9 show that the percent dye removal was increased up to an adsorbent dosage of about 4 g/L, after which it remained almost constant. Equilibrium was found to be attained more rapidly at low dye concentrations.

When excess adsorbent dosage is used, a significant portion of the adsorption sites remain unsaturated. This obviously leads to low specific adsorption efficiency, with

respect to usage of the substrate. When adsorbent dosage is lowered, the number of active sites saturated with dye increases; therefore, specific uptake also increases. Similar findings were reported by Ho and Ofomaja (2006) as well as Sharma *et al.* (2009).

### Solution pH

The pH variable is so important relative to adsorption of dyes that it has been given its own column in Table A. By examination of the table, some general trends can be observed. Entries in the first part of the table, devoted to basic dyes, generally show favorable results at relatively high pH values. By contrast, as shown the sections of the table dealing with acidic, direct, and reactive (*i.e.*, anionic) dyes, it is clear that negatively charged species were generally removed more effectively when the pH was very low. Both of these sets of results can be understood based on the dissociation of carboxylic acid groups on the substrates, which are often present predominantly on the hemicellulose and extractives components of lignocellulosic materials (Stenius and Laine 1994; Laine 1997; Lindgren *et al.* 2001, 2002). The  $pK_a$  values of such  $-COOH$  groups are typically in the range of 3.4 to 6, which implies increasingly negative charge as the pH rises within a range between about 2 and 8. Thus, in order to protonate all of the carboxylic acid functions on typical cellulose-related surfaces, thus removing the negative charge, it is necessary to reduce the pH to below 3. Accordingly, studies have shown that the highest sorption capacity of cellulosic materials for anionic dyes (acid, direct, or reactive) are often observed at pH values in a range between 1 and 3 (see, for instance, Etters and Hou 1991). Unfortunately, there has been little attention in the literature related to biosorption as to the presumed presence or identity of cationic sites at cellulosic surfaces at such low values of pH (Hu *et al.* 2010).

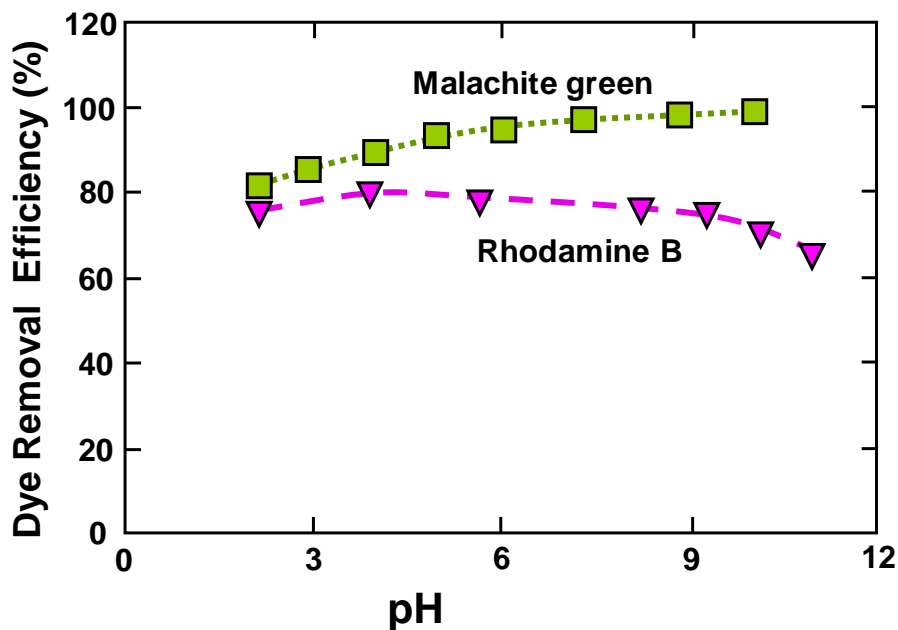
In general the role of pH can be understood by considering the influence of the  $H_3O^+$  and  $OH^-$  ions in regulating the electrical potential of cellulosic surfaces. Change of pH thus affects the adsorptive process through dissociation of functional groups on the adsorbent's surface-active sites. This subsequently leads to a shift in reaction kinetics and equilibrium characteristics of an adsorption process. At sufficiently low pH values, at which the sorbent surfaces have been protonated, negatively charged dye species are provided with a electrostatic driving force to migrate toward surface sites and become adsorbed. Conversely, at sufficiently high pH values, where the sorbent surfaces have interacted with hydroxyl ions, dye cations are adsorbed due to negatively charged surface sites of adsorbents. These relationships can be represented as follows:



In these schemes the dash (—) represents a transient chemical bond that may be considered as ionic or as a form of chemical complexation, depending on the case. The letters S denotes the adsorbent surface,  $A^-$  represents an anionic dye species, and  $C^+$  represents a cationic dye species.

The influence of pH on the surface characteristics of hydrated ion oxides and other solids have been investigated, for instance, by Janos *et al.* (2003) and Singh *et al.*

(2003). The adsorption of various cationic species on such adsorbents has been explained on the basis of the competitive adsorption of  $H^+$  and  $OH^-$  ions along with adsorbate molecules. For example (Fig. 10), the removal of malachite green (MG) was found to be favored at higher pH (Wang 2010). The best pH range for adsorption of MG was from 7 to 10. This result could be explained considering the electrostatic interaction between the surface of the adsorbent, which is negatively charged at sufficiently high pH, and the dye, which is present as a cationic species.



**Fig. 10.** Effect of solution pH on the adsorption of MG and RB by activated carbon ( $C_0=200$  mg/L; PLAC dosage=0.35 g/L; temperature= $25\pm 1$  °C;  $t$  (MG)=150 min,  $t$  (RB)=240 min); Data source (redrawn): Wang *et al.* (2010).

The variation in the uptake of MG dye with change in pH could be explained with reference to the zero point charge ( $pH_{zpc}$ ) of the material, which is the solution pH at which the adsorbent exhibits a net zero surface charge in the absence of any specific adsorption. Thus at  $pH < pH_{zpc}$ , the surfaces of adsorbents tend to be positively charged due to adsorption of  $H^+$  ions. Amine groups become protonated at pH below  $pH_{zpc}$ . At yet lower pH, carboxyl groups become protonated to their respective neutral forms. A force of repulsion develops between the dye cations and the protonated adsorbent surfaces. In addition, at lower pH the concentration of  $H^+$  ions is high, and they compete with positively charged MG cations for vacant adsorption sites. Thus at low pH, adsorption of MG dye was lower.

An opposite situation was observed at  $pH > pH_{zpc}$ , where the surface of adsorbents becomes negatively charged due to adsorption of  $OH^-$ ; the carboxyl groups of the adsorbents become deprotonated, producing negatively charged adsorption sites. Thus at high pH, adsorption of MG is highly favoured through the electrostatic forces of attraction. Similar trends were observed by Kumar and Porkodi (2007), Bulut and Aydin

(2006), and Kumar and Kumaran (2005) during adsorption of basic dye on different adsorbents. The maximum removal of MG was observed to be at pH 10. These results are generally in agreement with other reports (Namasivayam *et al.* 2001; Aksu 2001; Kannan and Sundaram 2001; Batzias and Sidiras 2007; Annaduria *et al.* 2002).

Though rhodamine B (RB) is usually regarded as a cationic dye, it showed a reverse trend in Fig. 10. Dye adsorption was greater in the acidic range. When  $\text{pH} < \text{pH}_{\text{zpc}}$  the adsorbent surfaces are positively charged. When  $\text{pH} > \text{pH}_{\text{zpc}}$ , adsorbent surfaces have negative charge due to dissociation of carboxylic group present in the anionic dye molecule, causing repulsion between negatively charged dye molecules and adsorbent surfaces, decreasing the adsorption of acid dyes in alkaline pH. Similar findings were obtained by Gupta *et al.* (2000). MG tended to be more adsorbable than RB. This may be due to its smaller ionic radius and lighter molecular weight. Tamai *et al.* (1999) reported that pore size is more important than the surface area as determined by the Brunauer, Emmett, Teller (BET) method. As explained by Walker and Weatherly (2001), only 14% of the BET surface area of commercial activated carbon (FS400) was available for dye adsorption. Besides molecular size, differences in molecular structure of these two dyes may be another reason for the variation in the dye adsorption capacity shown in Fig. 10.

### Ionic Strength

The ionic strength of an aqueous solution is a function the combined concentrations of the components ions (Hiemenz and Rajagopalan 1997). On the one hand, the ionic strength governs the distance over which charge effects can have significant influence; in distilled water an individual ion in solution will be attracted or repelled by charged groups that are many nanometers distant, whereas with increasing salinity, charge interactions are restricted to a much narrower zone. On the other hand, salt ions can be expected to compete with ions of the same sign of charge, including charged dye molecules, for adsorption sites. Because the salt ions may be much more numerous, one might expect dye adsorption to be adversely affected. However, there is a third important influence. The ionic strength of the solution also may affect the degree of solubility of a dye in the solution (Fedoseeva *et al.* 2010).

Diminished adsorption of dye with increasing salinity has been observed when cationic dye species are being adsorbed on cellulosic materials (Maurya *et al.* 2006; Han *et al.* 2007b; Cheng *et al.* 2008; Maurya and Mittal 2009; Zksu and Balibek 2010). This trend is consistent with the above-mentioned competition between cationic metal ion species and cationic dye molecules for negatively charged sites on the substrate. However, some other authors have reported cases in which increasing salt enhanced adsorption of basic dyes (Batzias and Sidiras 2007c; Dogan *et al.* 2008; Hu *et al.* 2010).

In the case of anionic dyes, increasing ionic strength generally has been found to favor adsorption onto cellulose-related substrates (Thode *et al.* 1952, 1953; Maekawa and Kondo 1996; Maekawa and Ohmori 1997; Ibbett *et al.* 2006b, 2007a; Bae *et al.* 1997a; Aksu and Balibek 2010). Bae *et al.* (1997b) observed that the presence of salt aided in the diffusion of anionic dye into a cellulose film substrate. In fact, certain procedures for the dyeing of cellulosic fabric material call for the use of saline solutions (Carillo *et al.* 2002). However, if the salt level is sufficiently high, evidence of competition between salt and reactive dyes for adsorption sites has been observed (Akar *et al.* 2009f).



## Surfactants

Because both surfactants and typical dyestuffs contain both hydrophobic and hydrophilic groups, it is reasonable to expect there to be some interesting and complex interactions between the two classes of material in solution. Aksu *et al.* (2010) observed that the adsorption of basic dye was greatly enhanced when an anionic surfactant was added to the solution. Iamazaki and Atvars (2007) reported beneficial effects of surfactant on adsorption of fluorescent whitening agent onto modified cellulose, but the effects were different depending on whether the surfactant was anionic or cationic. The beneficial effect of the cationic surfactants was observed below the critical micelle concentration (CMC). By contrast, the beneficial effect of anionic surfactant was observed only when it was present at sufficient concentration to exceed its CMC value. Moater *et al.* (2008) carried out an analysis to clarify the multiple interactions involved in the case of an anionic surfactant and anionic dye adsorbing onto anionic cotton. The authors concluded that adsorption of dye-surfactant complexes played a prominent role. Oakes and Dixon (2003) reported cases in which the presence of surfactant inhibited dye sorption, presumably by increasing the stability of dye species in the bulk or micellar phase.

Researchers have utilized cationic surfactants as a means of modifying cellulosic substrates and rendering them more capable of taking up anionic dye species (Oei *et al.* 2009; Akaer and Divriklioglu 2010; Akar *et al.* 2010).

## MECHANISTIC EXPLANATIONS FOR DYE REMOVAL

This section will discuss various explanations that have been proposed in the literature to account for the amounts of different dyes adsorbed on cellulose-related substrates. Topics are organized in the following main categories: thermodynamics, kinetics, and then consideration of the kinds of forces that may be responsible for drawing the dye molecule towards a site on the substrate.

An important point to keep in mind, when considering the discussions below, is that there is great diversity among both dyestuffs and cellulosic substrates. It is reasonable to expect, therefore, that the governing mechanisms may differ for different systems.

### Thermodynamics and the Effect of Temperature

A large amount of research has been carried out to answer questions such as, “Is heat generated in the process of adsorption?” Answers to this question can be found by inspecting two important columns of information in Table A. The column headed by “Best temperature” indicates whether the highest adsorption of the dye was observed at high, low, or at a specified temperature. Values in parentheses merely indicate the default temperature at which measurements were made; but the remaining values indicate the temperature at which the highest uptake was observed. The column headed by “Thermodynamics” indicates whether the data indicated an exothermic or an endothermic process, *i.e.* whether heat was generated during the process of adsorption.

Without exception, every study that considered the issue reported that adsorption of dye onto the substrate took place spontaneously, *i.e.* that there was a negative change in Gibbs free energy associated with the adsorption process. This is not a surprising conclusion, given the fact that every one of the studies was able to detect at least some finite adsorption of the dye. What is more interesting in Table A is the fact that there was an almost equal representation of systems that were found to be endothermic vs. exothermic. In this context the word “exothermic” implies that the net change in heat related to the forces of interaction among the molecules favors adsorption. Generally there are no covalent bonds created or destroyed during adsorption of a dye onto a cellulosic substrate; rather the change in enthalpy is associated mainly with electrostatic and van der Waals attractions among the component molecules.

Further inspection of Table A will reveal that all of the systems listed as being endothermic in character exhibited highest adsorption at the high end of the temperature range considered by the investigators. By contrast, all cases that were judged to represent exothermic adsorption were maximized at the low end of the temperature range tested. These relationships follow from the thermodynamic rule,

$$\Delta G = \Delta H - T\Delta S \quad (4)$$

where  $\Delta G$  is the change in Gibbs free energy during the process,  $\Delta H$  is the corresponding change in enthalpy or “heat,”  $T$  is the absolute temperature, and  $\Delta S$  is the change in entropy. Entropy is a measure of the disorderliness of the system. In other words if the adsorption of a dye molecule on a substrate has a net result of there being a larger number of individual molecules able to move around with greater freedom, then such a change will tend to contribute to the process going forward.

It is important to make a cautionary statement, however: because the adsorption of a dye molecule can either increase or decrease the degree of orderliness (or “structure”) of the surrounding water molecules, changes in entropy are not easy to predict theoretically. Fortunately, the experimental approach to determining whether a process takes place endothermically or exothermically follows from the form of Equation 5. One assumes, first of all, that the nature of the process remains basically the same throughout the temperature range of investigation, *i.e.* one assumes that the amount of heat associated with the physical forces responsible for holding the dye molecule on the substrate does not change. Tests are then carried out at different temperatures. Most investigators whose results are listed in Table A chose to employ three different temperatures; by such means they can demonstrate a linear relationship between adsorption and absolute temperature, consistent with theory. The important result, observed from many such investigations, is that effective adsorption of dye does not require that heat is generated in the process; rather, it becomes clear from the results of many studies that entropy considerations play a dominant role in the adsorption of typical dyes onto cellulose-related substrates.

In practical terms the temperature plays a vital role in adsorption processes. On the one hand, increasing the temperature results in increasing the rate of diffusion of the adsorbate molecules across the external boundary layer and in the internal pores of the adsorbent particles. On the other hand, the equilibrium capacity of the adsorbent for a

particular adsorbate may change (Al-Qodah 2000). The latter effect may be related to a change in the dye's solubility; an inverse relation between the adsorbate solubility and adsorption capacity has been observed in many cases. Less soluble materials tend to get adsorbed preferably, and the more soluble materials do so less readily (Asfour *et al.* 1985). Contradictory observations, however, have been made by the previous worker regarding the effects of temperature on the rate of adsorption. Several earlier investigators (Ho and McKay 1999; Karagoz *et al.* 2008; Tunc *et al.* 2009; Jain and Sikarwar 2009) reported an increase in uptake of adsorbate at higher temperature, which may be explained by carefully examining the mode and type of adsorption processes. Diffusion of adsorbate species from the bulk phase into the pores of adsorbent has been found to be important in some of the uptake processes that are of endothermic character; *i.e.* increasing temperature favors the adsorbate transport within the pores of the adsorbent. Karaca *et al.* (2006) explained the increased adsorption with rise of temperature on the basis of ion exchange reactions between adsorbate and adsorbent. The increase in adsorption with temperature is also attributed to the breaking of some of the internal bonds near the edge of the active surface site of adsorbents. Efforts have also been made to explain the increase or decrease in adsorption with change in temperature on the basis of variation in chemical potential of the system (Chandra *et al.* 2007). Contradictory observations have been made by the several workers regarding the effect of temperature on the rate of adsorption (Ozer and Dursun 2007; Gercel *et al.* 2007; Mane *et al.* 2007; Gupta *et al.* 2003; Sharma *et al.* 2009). The latter authors reported increases in uptake of adsorbate with increase in temperature, *i.e.* endothermic adsorption.

Adsorption studies are often carried out at different temperature, keeping the concentration, pH, and other parameters constant throughout. Though the results of such tests usually are interpreted in terms of the endothermic or exothermic nature of the process, other explanations are possible. For instance, increased adsorption with increasing temperature may be due to an increase in the mobility of the dye (Dogan and Alkan 2000), making it possible to come closer to a saturated condition within the time of experimentation. Also, with increasing temperature, a greater number of molecules may acquire sufficient energy to surpass an energy barrier involved in the interaction with an active site on the adsorbent surfaces. Furthermore, increasing temperature may produce a swelling effect within the internal structure of the activated adsorbents, enabling a large dye molecule to penetrate further (Asfour *et al.* 1985). Thus it has been suggested that an increase of adsorption capacity of the adsorbent at higher temperature may be due to enlargement of pore size and activation of the adsorbent surface (Weber 1967).

### Kinetic Expressions: Effects of Time on Amount of Dye Adsorbed

Essentially all investigators, if they report anything at all regarding kinetics, have reported that the amount of dye adsorbed onto a cellulose-based substrate increases with increasing time in contact with the solution. More important questions are, (1) what is the rate law governing such adsorption?, and (2) what mechanism can account for that rate law?

Answers to the first question can be seen in Table A under the heading "Rate law best fit." With just a few exceptions, most investigators who have reported rate laws have found best fits to a so-called "pseudo-second-order" rate expression (Ho and McKay

1998a-c, 1999; McKay and Ho 1999). In this term the prefix “pseudo” implies that though the true rate law governing the interaction might possibly be more complex, within the range of variables considered it is possible to fit the data to an expression of the following form (Ho and McKay 1998d, 1999),

$$dq_t / dt = k (q_e - q_t)^2 \quad (5)$$

where  $q_t$  is the amount of dye adsorbed at time  $t$ ,  $k$  is the rate constant, and  $q_e$  is the amount adsorbed at equilibrium. This expression can be integrated to yield,

$$1 / (q_e - q_t) = (1 / q_e) + kt \quad (6)$$

which can be rearranged as:

$$(t / q_t) = 1/(k_2 q_e^2) + (1/q_e)t \quad (7)$$

When adsorption data fit well to a rate expression of this type, one possible explanation has been that the rate-determining step of adsorption involves participation of two adjacent functional groups at the substrate surface. By contrast, if data were to fit a first-order rate expression, consistent with some other cases listed in Table A, then one might conclude that the rate-limiting step involves adsorption of a solute molecule on an individual site on the substrate. Such mechanisms might be considered in cases where there is clear evidence that adsorption is correlated to certain functional groups on the substrates; however the present reviewing effort did not discover any attempt to justify the use of a pseudo-second-order model to explain the rates of adsorption of dyes by such a mechanism.

Another way to interpret the prevalence of excellent fits to the pseudo-second-order rate model is to observe the practical implications of the form of Eq. 5. The rate is shown to depend on the square of the “unfilled” adsorption sites. That means that even if the equation might have been derived with another mechanism in mind, its practical effect is to help fit data in which the last remaining sites become increasingly reluctant to be filled. A number of mechanistic reasons for such reluctance can be listed, *e.g.* charge repulsion between like-charged dye molecules, physical crowding between the dye molecules as the saturated condition of the substrate is approached, and also the possibility that certain sites are simply not as favorable. This last possibility – the existence of less favorable sites – is especially interesting to consider in cases where dyes are capable of permeating within the cell walls of fibers. As noted in an earlier review article, one can expect a slowing of rates of sorption if adsorbate molecules are required to proceed “single file” to get into the interior of a mesoporous material (Wu *et al.* 2009).

#### *Diffusion into the cell wall*

Most researchers who have expressed an opinion on the topic have agreed that the rate-limiting step for adsorption of dye onto cellulose-related substrates must involve diffusion, though it is not fully resolved whether such diffusion resembles bulk diffusion or if it takes place essentially within a system of small pores (Morita *et al.* 1985, 1986;

Maekawa *et al.* 1989; Maekawa and Kondo 1996; Petzold *et al.* 1997; Bhattacharyya and Sharma 2005; Özer *et al.* 2006; Porkodi and Kumar 2007; Vijayaraghavan *et al.* 2007). Some authors have employed a “boundary layer” diffusion model, in which one envisions that the solute needs to pass through a hypothetical layer that offers some resistance to transport before it can become adsorbed (Akkaya and Ozer 2005; Li *et al.* 2010).

An intra-particle diffusion model (Weber and Morris 1963) has been supported by various research (Waranusantigul *et al.* 2003; Suteu and Bilba 2005; Vijayaraghavan and Yn 2007a; Mohan *et al.* 2008; Ofomaja 2008; Laohaprapanon *et al.* 2010; Nethaji *et al.* 2010; Rubin *et al.* 2010; Thirumalisamy and Subbian 2010). In this approach the process is divided into two parts. The first step is presumed to involve adsorption at an exterior surface or boundary layer of the substrate particles. Presumably this step can occur rapidly relative to other processes. The second step involves permeation by diffusion into pore sites, a process that is presumed to depend on the square-root of time. The combined process has been proposed to follow a rate law given by (Yang *et al.* 2011b),

$$Q = k t^{0.5} + I \quad (8)$$

where  $Q$  is the amount adsorbed per unit mass of sorbent,  $k$  is the rate constant governing diffusion into the particle,  $t$  is the elapsed time after combining the solution and the sorbate, and  $I$  is the intercept of a plot of  $Q$  versus  $t^{0.5}$ . In cases where the plot goes through the origin, one has evidence that the boundary effect can be neglected. Other researchers have found evidence of effects due to both diffusion from the bulk of solution and diffusion within the substrate (Dizge *et al.* 2008). Tseng *et al.* (2011) demonstrated the use of a model involving the progressive filling of adsorbable sites within a porous structure; such a model was able to explain the rates of uptake of methylene blue dye on an activated carbon substrate. Thus, the value of  $I$  in Eq. 8 can represent the amount of dye that can quickly saturate the external sites of adsorption. Issues related to diffusion into the pore spaces of biomaterials are considered in greater detail in Wu *et al.* (2009).

A minority of researchers have actually attempted to measure the concentration-distance profiles within cellulosic materials as a means of quantifying rates of diffusion of a dye within the solid substrate. Bae *et al.* (1997b) were able to carry out such an analysis by preparing a tightly wrapped cylinder of cellulose film; after a set period of adsorption, the film was quickly uncoiled so that the different sections – representing different distances from the surface – could be evaluated for dye content. Sarkar and Bandyopadhyay (2010) likewise showed that a “shrinking core” model could be fit well to dye adsorption data for basic dyes onto rice husk ash.

### Agitation

Whether or not diffusion in the bulk of solution plays a significant role with respect to dye adsorption is likely to depend on whether or not the solution is stirred. A number of researchers have explicitly included agitation as an independent variable. Batzias and Sidiras (2007) reported an increase in rate of dye removal with increased degree of agitation for the adsorption of methylene blue on beech saw dust. This was attributed to the decrease, with increasing speed of agitation, in the diffusion boundary layer thickness surrounding the adsorbent particles. Adb El-Rahim and El-Ardy (2011)

found, surprisingly, that higher uptake of Direct Violet dye was obtained on fungal biomass in the absence of agitation. Allen *et al.* (2005) observed only very minor increases in uptake of basic dye by kudzu with increasing rates of agitation. Likewise, Asma *et al.* (2006) found only slight increases in adsorbed amounts of reactive dyes on fungal biomass with increasing agitation. Others achieved higher adsorbed amounts earlier in the presence of agitation (Sadhasivam *et al.* 2007a; Ponnusami *et al.* 2009; Wang *et al.* 2009; Rajeshkannan *et al.* 2010). In summary, transport of dye from the bulk of solution to the substrate surface does not seem to be a highly critical issue, according to most researchers.

### Charge Interactions

Irrespective of how one explains the thermodynamics and kinetics of dye adsorption, a fuller description of the mechanisms should also include forces of interaction. In principle, such forces can be described as being either electrostatic, covalent, hydrogen bonding, or as falling within various categories of van der Waals forces (Dzyaloshinskii *et al.* 1961; Visser 1972, 1995; Bowen and Jenner 1995), such as dispersion forces, dipolar interactions, acid-base associations, co-ordinate bonds, *etc.*

The clearest evidence that charge plays a major role in dye adsorption comes from the effects of pH. As was noted earlier, Table A shows that numerous studies involving basic dyes reported highest adsorbed amounts when the pH was relatively high, such that the carboxylic acid groups on the substrate surfaces would have been in their dissociated, negatively charged form. By contrast, anionic dyes generally exhibited higher adsorption when the pH was relatively low, conditions that suppress the negative charge of cellulose-related materials.

Further evidence of the importance of electrostatic forces comes from many studies that have used pH changes as a means of desorbing dyes, making it possible to regenerate the substrate and use it again (Laszlo 1995; Won *et al.* 2006; Vijayaraghavan and Yun 2007a,b, 2008c,d; Vijayaraghavan *et al.* 2007, 2008a,b; Patel and Suresh 2008; Mao *et al.* 2008, 2009a,b; Jain *et al.* 2010). In these studies the pH was adjusted in such a way as to render charge-charge interactions unfavorable for adsorption.

### Functional Groups

Various researchers have presented evidence in favor of the involvement of particular functional groups at cellulose-related surfaces playing an important role in the adsorption of dyes. In many cases, such evidence has consisted of shifts in the wavelengths of absorbance maxima in infra-red spectra upon adsorption of a dye (Odochian *et al.* 2007; Das *et al.* 2008; Zafar *et al.* 2008; Akar *et al.* 2009b; Saeed *et al.* 2009; Won *et al.* 2009c). Some caution is required when interpreting such effects. A charged dye molecule, just by its presence, can be expected to alter the electron densities adjacent to various functional groups on the substrate surface. As a consequence, the spring constants of various covalent bonds within the molecules can be shifted. Such effects can be expected to influence the infra-red adsorption characteristics regardless of whether the functional group has a significant influence on the adsorption.

In other cases infra-red measurements have been used simply as a tool to show the presence of functional groups, which presumably would be available for interactions with

sorbates (Maurya and Mittal 2008; Alam *et al.* 2009; Mao *et al.* 2009c). Thus, Bangash and Alam (2007) were able to demonstrate the disappearance of surface carboxyl, lactone, ether, and hydroxyl groups upon high-temperature activation of carbon. In this case the adsorption of dye was favored by the activation, and presumably by the lowered amount of oxygen-containing groups. Nasr *et al.* (2006) studied a case in which the presence of carboxyl groups on activated carbons favored adsorption of cationic dye. Peterlin *et al.* (2009a) found a similar trend when comparing the uptake of cationic dyes onto thermomechanical pulp fibers that had been oxidized to different degrees.

A more persuasive demonstration of the importance of certain functional groups was achieved by Gong *et al.* (2005), who selectively modified amino, carboxyl, and hydroxyl groups of peanut hull biomass, thereby eliminating the ionic charge, and monitored the adsorption of a variety of dyes. Carboxyl groups were thus shown to play a major role in the observed dye adsorption. Not only were the carboxyl groups shown to favor the adsorption of basic dyes, but they also were shown to interfere with the adsorption of various anionic dyes. Similar results were obtained by Han and Yun (2007). Hydroxyl groups were found to be important for adsorption of all six dyes that were considered by Gong *et al.* (2005); the latter effect can provide additional support for hypotheses that hydrogen bonding can play an important role in dye sorption.

Some of the highest-performing sorbent materials, still based on cellulosic materials, have been prepared by chemical derivatization or grafting reactions capable of achieving a high density of selected functional groups on the substrate. The effectiveness of such preparations can provide further evidence of the role of such groups with respect to the uptake of dyes. For example, Hwang and Chen (1993a,b) showed greatly enhanced sorption of direct dyes (anionic) onto cellulose that had been modified with a combination of adipic acid, cationic polymers, and cross-linking agent.

### Chemical Complexation

Metal ions appear to play a central role in the adsorption of certain textile dyes. Such systems can be described as chemical complexes, in which the dye molecules function as chelating agents (Lawrence 2010). Lima *et al.* (2008) showed that treatment of biomass with chromium increased its ability to sorb reactive dyes. Likewise Saliba *et al.* (2002) achieved increased uptake of various dyes by co-adsorption of various divalent or trivalent metal ions. Such complexes appeared to be heat-sensitive, such that it was possible to totally desorb the dyes from the substrate by heating the co-precipitated mixture. Baouab *et al.* (2007) demonstrated such a mechanism by using a spectrometric analysis of the various organo-metallic complex species.

It is well known that the affinity of various anionic dyes for papermaking fibers can be increased by addition of calcium or aluminum cationic species (Lips 1981; Galafassi and Paciello 1985; Hubbe *et al.* 2008). Togo and Komake (2010) described the use of aluminum salts as fixatives for tannic acid species, as a means of using a natural colorant for cotton. In such applications the metal ions appear to act as a mordant, serving as a kind of bridge between the negative charges of the substrate and various anionic groups associated with the chromophoric molecules.

### Hydrophobic Association and Pi-Pi Association

The association between hydrophobic parts of dye molecules with various substrates has been widely discussed as a possible contribution to adsorption (Kuhn and Lederer 1988; Bami-Horn *et al.* 1992; Timofei *et al.* 1994; Oakes and Dixon 2003; Abbott *et al.* 2004a; Elizalde-Gonzalez and Hernandez-Montoya 2009b). Hamlin and Whiting (2005) used molecular dynamics simulation to demonstrate the tendency for dyes to self-associate, an effect that was attributed to their content of aromatic groups. Abbott *et al.* (2004a) found evidence that a direct dye associates itself with apolar regions of the cellulose surface, thus supporting a mechanism in which hydrophobic effects contribute to bonding. It is reasonable to expect that London dispersion forces among the apolar molecular groups, in conjunction with a tendency for association among hydrophilic groups in the system, provides the basis for these interactions.

A related effect was demonstrated during the adsorption of relatively high amounts of fluorescent whitening agent onto cellulose fibers (Stana *et al.* 1995). Beyond a certain concentration the sulfonated molecules appeared to adsorb in multi-layer fashion. The change from monolayer to multilayer adsorption was evident from spectral changes. By contrast, Yamaki *et al.* (2005) did not detect any spectral or other evidence of dye self-association during layer-by-layer preparation of dye-modified cellulose fibers.

The planar structure of various dye molecules appears to favor a stacking of molecules on top of each other such that aromatic groups are aligned (Pielesz 2007). Aspland and Bird (1961) provided early evidence to support such a mechanism in the case of non-ionic dyes and their interaction with viscose rayon. It was shown, for instance, that a co-planar structure of the sorbate molecule led to a higher level of sorption. The effect, which has been called pi-pi stacking, has been used extensively in development of dye technologies, for instance in the preparation of laked pigment dyes (Chakraborty 2010). Pielesz (2007) used NMR methods to demonstrate pi-pi stacking of direct dyes, in addition to hydrogen bonding of the associated dyes with a cellulose substrate.

One of the promising ways to demonstrate association among dye molecules, as a part of an adsorption mechanism, is by measurement of fluorescence effects. Zacharova and Chibisov (2008) used fluorescence effects to show changes in the association between thiacyanine dye molecules upon their adsorption onto cellulose. Adsorption increased the proportion of dimerization between the dye molecules.

As noted by Maekawa and Kondo (1996), aggregation of dye molecules has the potential to hinder transport within the fine pores of various substrates, where presumably individual dye molecules would more easily permeate.

### Hydrogen Bonding to the Substrate and Molecular Orientation

Some of the best evidence for the participation of hydrogen bonds in the binding of dyes to cellulose has been obtained with Raman spectroscopy (Abbott *et al.* 2004a). The cited authors showed that when Direct Blue 1 was dissolved in organic solvent there was internally-directed hydrogen bonding within the dye molecules. In aqueous solution, by contrast, these interactions were directed outwards, toward the solvent. And in the adsorbed state, there was significant hydrogen bonding with cellulose. Al-Ghouti *et al.*



(2010) used infra-red analysis to find evidence of hydrogen bonding between methylene blue and date pit biomass.

Various advances in understanding of dye adsorption mechanisms have been achieved by means of mathematical and computer simulation methods. Timofei *et al.* (1994, 2000, 2001) found a good correlation between the affinity of different dyes and their planar conformations. Likewise Bird *et al.* (2006) correlated dye structure and dye uptake data and concluded that the highest binding enthalpy could be achieved with dyes that had their solubilizing groups (sulfonates) all facing in one direction, making it possible to maximize binding enthalpy when the other side faces the substrate. According to each of these analyses the orientation of dyes during adsorption appears to be important.

### Chemisorption vs. Physisorption

The question of whether dye sorption should be considered as chemisorption vs. physisorption has been discussed in a number of studies, among which there appears to be little agreement (Wardas and Lebek 1994; Namasivaham and Kavitha 2002; Binupriya *et al.* 2007; Tarawou *et al.* 2007; van de Ven *et al.* 2007; Maurya and Mittal 2009; Tan *et al.* 2010). Namasivaham and Kavitha (2002) and Senthikumaar *et al.* (2006a) judged that the observed adsorption should be considered as chemisorption because very little of the dye could be removed, even when the pH of the rinsing solution was varied in a wide range. Various researchers have used the term chemisorption to indicate that the adsorption or desorption process was influenced by the pH and ionic charges in the system (Binupriya *et al.* 2007, 2008b; Kavitha and Namasivayam 2007a; Sathishkumar *et al.* 2007). Other authors cite pseudo-second-order adsorption kinetics as evidence of chemisorption (Senthikumaar *et al.* 2006b; Ncibi *et al.* 2008a, 2009c; Xing and Deng 2009; Suteu *et al.* 2010). Won *et al.* (2008) used the term chemisorption to describe the reaction of a reactive dye, rendering it difficult to remove from a substrate.

The term physisorption, likewise, has been employed in various ways. Bekci *et al.* (2009) used the values and sign of free energies of adsorption as grounds for characterizing malachite green adsorption on algae as a case of physisorption. van de Ven *et al.* (2007) used the term to indicate interactions that did not involve electrostatic effects. Palanisamy and Sivakumar (2009) used a slow rate of adsorption as their criterion for classifying adsorption of Acid Blue 92 on biomass as a case of physisorption.

There appears to be a lack of common usage for the terms chemisorption or physisorption, at least within the context of dye bioremediation. As such, it is recommended that future authors who use such terms be careful to provide working definitions of key terms.

### What the Shapes of Sorption Isotherms May Tell Us

#### *The Langmuir isotherm*

Notations in Table A show that the various different models have been used successfully to fit adsorption isotherm data for dyes onto various cellulosic substrates, but that by far the most widely used model has been that of Langmuir (1918). Most authors have interpreted accurate fits to the Langmuir equation by drawing conclusions that (a) the substrate has a specified capacity for adsorption of the dye, and (b) the adsorption

sites are equivalent in terms of energy. Though many authors have used the term “monolayer” to explain the implications of a good fit to the Langmuir equation, few studies have provided evidence or claimed that the dye adsorption was limited to external surfaces. Rather, the Langmuir equations seems to have widespread applicability in cases where the dye is able to diffuse into the cell walls of lignocellulosic material. For instance, the following researchers reported good fits to a Langmuir isotherm, while at the same time reporting that the rate of adsorption was at least partly governed by diffusion into the cellulosic material (Aretxaga *et al.* 2001; Malik 2003; Waranusantigul *et al.* 2003; Mahdaoui 2005; Senthikumaar *et al.* 2005; Vadivelan and Vasanth Kumar 2005; Vasanth Kumar *et al.* 2005; Aksu and Isoglu 2006; Gulhaz *et al.* 2006; Gercel *et al.* 2007; Kavitha and Namasivayam 2007a; Porkodi and Kumar 2007; Dizge *et al.* 2008; Hameed and El-Khaiary 2008a,b; Onal *et al.* 2007; Vijayaraghavan *et al.* 2007, 2008b; Aksu *et al.* 2008; Aksu and Karabayir 2008; Mohan *et al.* 2008; Alzaydien 2009; Panda *et al.* 2009; Samiey and Dargahi 2010; Akar *et al.* 2010; Yang 2011b). Further evidence in support of a three-dimensional model of dye sorption (either in mesopores or otherwise diffusing into the solid) includes the extensive time required for equilibration of dye solutions with cellulosic substrates, and also the relatively high amounts of dye adsorbed per unit mass of solid (see Table A). In typical cases it takes 20 minutes to more than an hour to reach a plateau level of adsorption (see, for example, Ouazene and Sahmoune 2010). So in summary, while a good fit to the Langmuir equation can suggest that there is a finite adsorption capacity, and that sites are energetically equivalent (and non-interacting), at least as an approximation, one should be cautious in use of the term “monolayer,” which could lead to an incorrect qualitative impression of what happens at a molecular level.

#### *The Freundlich isotherm*

Table A also shows that a prominent minority of studies considered in this review reported good fits to the Freundlich equation (Freundlich 1907). Almost universally, authors of articles represented in Table A have concluded, based on such fits, that the substrate contained a diversity of sorption sites, having different affinities towards the target dye material. In the case of relatively weak binding sites, one can expect there to be a strong dependence of the adsorbed amount on the solution concentration, even at relatively high values of bulk concentration.

Due to their charged nature it is reasonable to expect that many dye chemicals will adsorb preferentially, with higher energy, at sites of opposite charge on cellulosic surfaces; however it is reasonable to expect that the same dye molecules can adsorb, albeit with weaker affinity, due to hydrophobic interactions at uncharged sites on the substrate. Further investigation would be needed in order to demonstrate whether this is a useful concept.

#### *Fits simultaneously to more than one isotherm model*

An interesting question can arise in cases where authors have reported good fits to two or more isotherm models. For instance, several authors have reported simultaneously good fits to both the Langmuir and the Freundlich equations (Annandurai *et al.* 2002; Namasivayam and Kavitha 2002; Malik 2003; Banerjee and Dastidar 2005;

Bhattacharyya and Sharma 2005; Gong *et al.* 2005; Eren and Acar 2006; Hashem 2006; Nacera and Aicha 2006; Nasr *et al.* 2006; Padmesh *et al.* 2006a; Preethi *et al.* 2006; Ahmade *et al.* 2007; Kumari and Abraham 2007; Porkodi and Kumar 2007; Sadhasivam *et al.* 2007a,b; Al-Degs *et al.* 2008; Cheng *et al.* 2008; Dizge *et al.* 2008; Lata *et al.* 2008c; Low *et al.* 2008; Oliveira *et al.* 2008; Vijayaraghavan and Yun 2008c; Gad and El-Sayed 2009; Panda *et al.* 2009; Anjaneya *et al.* 2009; Janoš *et al.* 2009; Sekhar *et al.* 2009; Aksakal and Uzun 2010; Gao *et al.* 2010; Lim *et al.* 2010; Xiong *et al.* 2010; Siroky *et al.* 2011; Sivasamy and Sundarabalan 2011). In some such cases it is likely that a better discrimination between the competing models could have been achieved by extending the data range to higher equilibrium concentrations. Also, one can argue in many cases that there was insufficient statistical evidence to rule out either model as providing an adequate description of the data. In the latter cases it makes sense to favor the Langmuir model. The Langmuir equation is based on a more concrete mechanistic model in comparison with the Freundlich equation, which can be considered as a way to summarize empirical data. A further reason to prefer the Langmuir equation is that one can readily compare adsorption capacities obtained in different studies. Porter (1993, 2002) also showed that the Langmuir model can be modified to account for specific interactions during competitive adsorption of two different dyes and sites on the same substrate. Indeed already since 1974 (Johnson *et al.* 1974) there has been various evidence that certain dyes compete for the same adsorption sites, whereas other pairs of dyes seem to adsorb more independently of each other. More research attention, however, is needed in order to help understand why this is so.

## ISSUES NEEDING FURTHER RESEARCH

As is evident from the studies represented in Table A, many cellulose-derived materials have been shown to be quite effective for the removal of dyestuffs from aqueous solutions. Furthermore, many of the cited studies have demonstrated ways to modify the biosorbents to improve their performance, and it is also apparent that results can be improved by judicious choice of low-cost sorbent material. Challenges remain, however, when it comes to practical implementation of biosorption. While further studies of the capacities of different cellulose-related substrates to remove dyes from aqueous systems will continue to have value, the results of this review of the literature suggest that the most urgent priorities lie elsewhere. There is a great need for additional research concerning how to further process or dispose of the biosorbent after it has been used to collect dyes, some of which may be toxic, slow to biodegrade, or subject to leaching. This section will review various studies that shed light on such questions as “what does one do with the biosorbent after it has been used?”.

Some key areas that appear to have received inadequate attention include the following:

- Feasibility and costs associated with collection, transportation, drying, preparation, and storage of proposed substrates for adsorption of dyes.

- Issues concerning the flow characteristics of packed beds of biomaterials, especially in the case of compressible materials such as bacterial or fungal biomass. Though immobilization of fines and compressible biomaterials onto more rigid and highly permeable substrates has been studied, there is a need for more research of a practical nature in this area.
- Life cycle and processing issues associated with possible regeneration of biosorbents after they have been used for removal of dyes.
- Environmental issues associated with dye-loaded cellulosic substrates, *e.g.* in landfill disposal, composting, or when used in the fabrication of secondary products.
- Issues related to incineration of wet, dye-containing cellulosic materials, *e.g.*, air emission quality and the degree to which energy is recovered from the incineration of biomass.
- Technology to increase the efficiency with which dye materials are retained during the manufacture of textiles, paper products, and other products so that potential impact on the environment can be avoided earlier in the process, before any wastewater has left the zone of primary manufacturing.

Table 4 lists some of the general approaches that might be considered, and it also lists some of the key positive and negative issues that have been associated with each of these approaches. Further details are discussed in the subsections that follow.

**Table 4.** Alternative Ways to Deal with Dye-Loaded Cellulosic Matter

Class of Approach	Positive attributes	Negative attributes
Incineration	<ul style="list-style-type: none"> <li>• Molecules are destroyed.</li> <li>• Energy is recovered (?)</li> </ul>	<ul style="list-style-type: none"> <li>• Wet biomass hard to burn.</li> <li>• Air pollutants formed.</li> <li>• Salty water a problem.</li> </ul>
Regeneration by desorption	<ul style="list-style-type: none"> <li>• Multiple use of sorbent.</li> <li>• Reduced shipping of solid.</li> </ul>	<ul style="list-style-type: none"> <li>• Need to treat the filtrate.</li> <li>• Strong acid or base reqd.</li> <li>• Salty water formed.</li> </ul>
Landfilling	<ul style="list-style-type: none"> <li>• Seeming simplicity.</li> </ul>	<ul style="list-style-type: none"> <li>• Big danger of leaching.</li> <li>• Lack of biodegradation.</li> <li>• Land must be condemned.</li> </ul>
Composting	<ul style="list-style-type: none"> <li>• Reduction in mass, toxicity.</li> </ul>	<ul style="list-style-type: none"> <li>• Product has no value.</li> </ul>
Wet enzymatic treatment	<ul style="list-style-type: none"> <li>• Destroys molecules.</li> <li>• Avoids harmful chemicals.</li> </ul>	<ul style="list-style-type: none"> <li>• Cost of enzymes.</li> <li>• Capital requirements.</li> <li>• Dye-specificity.</li> </ul>
Oxidation	<ul style="list-style-type: none"> <li>• Destroys molecules.</li> </ul>	<ul style="list-style-type: none"> <li>• Strong chemicals used.</li> <li>• Water treatment needed.</li> </ul>

### *Incineration*

Incineration can be considered to be a relatively mature technology, for which issues such as capital requirements, energy balances, and factors affecting the toxicity and particulate content of the exhaust gases have been well studied (Santoleri *et al.* 2000; Niessen 2010). According to Santoleri *et al.* (2000) incineration can be considered as the

most viable current technology for safe disposal of hazardous organic materials, which can be destroyed with high efficiency as long as sufficiently high temperatures are reached (*e.g.* 720 to 850 °C) under conditions allowing full oxidation. To ensure complete destruction of possibly hazardous organic materials, Neissen (2010) suggests the use of a rotary kiln operating above 950 °C, along with uniform mixing and a residence time of 1 to 2 seconds at the peak temperature. A gaseous content of 3% oxygen is recommended to achieve oxidizing conditions, keeping in mind that higher levels of oxygen, achieved by adding more air, may tend to lower the temperature of the system.

Consequences of the moisture content of the substrate are of particular concern here, since cellulosic materials typically can be expected to be wet after they have been used to collect dye chemicals from aqueous systems. Incineration processes are known to be affected by different levels of moisture in the feed material (Turovskii and Mathai 2006). The heating value of the material is reduced by moisture, and it becomes more difficult to achieve the high temperatures needed for full consumption of the feed material. It is well known that the amount of excess water can be reduced by various thickening options, such as by the use of a belt press or screw press (Turovskii and Mathai 2006; Wiesmann 2007). Filtrate from such pressing operations, which is likely to include some of the dye material in soluble or colloidal form, would need to be returned to an earlier stage of the wastewater treatment operation.

A few studies have considered aspects of incineration of residue from dyestuffs (see, for instance, Jang *et al.* 2001; Lee *et al.* 2001; Van der Bruggen *et al.* 2004; Yan *et al.* 2006). Jang *et al.* (2001) placed particular concern on NO<sub>x</sub> emissions, noting that the quantity of gaseous toxins can be highly dependent on operating parameters, such as the air-to-fuel ratio. However, studies focusing on the incineration of dye-containing cellulosic materials are rare (Ozoh 1997).

It should be noted that the term “incineration” has been appropriated, in a potentially confusing manner, to also cover certain electrochemical treatments, *i.e.* “electrochemical incineration” (see, for instance, Panizza and Cerisola 2008; Andrade *et al.* 2009; Zhao *et al.* 2009). Because such treatments involve oxidation taking place on the surface of an electrode, such as activated platinum, it is not obvious that the approach could be used for treatment of dyes adsorbed onto biomass.

#### *Regeneration by desorption*

Many of the biosorption studies cited in this review have demonstrated the possibility of regenerating the biomass by exposing it, for instance, to strongly basic or acidic pH. In general, very low pH conditions can be expected to bring about the desorption of basic dyes (Acemiglu *et al.* 2010; Yu *et al.* 2009a), whereas the converse can be expected for various negatively charged dyes (Bakshi *et al.* 2006; Zeroual *et al.* 2006b; Vijayaraghavn and Yun 2007a; Patel and Suresh 2008; Mao *et al.* 2009b). In theory, such regeneration would make it possible to utilize the biosorbent material multiple times. Ideally one would want to be able to regenerate the biosorbent material in place, as in the case of a packed bed treatment system (Vijayaraghavn and Yun 2008d).

Researchers have seldom addressed the question of what is to be done with the wash water after dye has been intentionally desorbed from a cellulosic substrate for

purposes of regeneration. Unlike the case of biosorption of metals, which was considered in an earlier review (Hubbe *et al.* 2011), it is considered unlikely that the material recovered from a biosorbent (the dyes) would ever be concentrated and purified for subsequent use. The recovered dyestuffs would tend to be unpredictable mixtures, having unpredictable behaviors and colors (Bisschops and Spanjers 2003). Moreover, the high or low pH used in the regeneration of the biosorbent would necessitate additional pH adjustment as part of any subsequent wastewater treatment system. Due to such concerns, regeneration of biosorbent by means of pH adjustment could be considered only in cases where facilities and treatment methods are in place to deal with the resulting dye-enriched solution, which may be either acidic, alkaline, or containing a relatively high salt concentration as a result of the pH adjustments. In addition to treatment of highly colored wastewater with a conventional activated sludge system (see introductory section of this article), it makes sense to employ such strategies as oxidative bleaching (Tozer 1993; Wang *et al.* 2006) and treatment with enzymes that catalyze breakdown of dyes (Anastasi *et al.* 2006; Rodríguez-Couto 2009; Papinutti and Forchiassin 2010).

### *Landfilling*

The option of hauling spent, dye-containing sorbent material to a landfill is sure to be considered, especially in cases where other options are judged to be too expensive. Whenever cellulosic products are landfilled, it is reasonable to expect the evolution of methane gas (Padgett 2009). Due to a lack of air circulation, in combination with the presence of various oxidizable substances, conditions within a landfill tend to become anaerobic, leading to the production of greenhouse gases. In state-of-the-art landfill facilities, provisions are made for the collection of the methane, which then can be used for fuel or other purposes (Kumar *et al.* 2011). However, there is further evidence that landfilling can be quite inefficient, if it is viewed as a process of biodegradation; there are accounts of investigators being able to read the pages of printed texts that had been buried in landfills for many years (Fogel 2002).

A major concern that merits continuing study is whether significant amounts of toxic or colored materials are likely to leach from such a landfill site and enter groundwater. Leachates from landfills already are a major source of concern, due to the potential for contamination of groundwater or surface water (Kuniawan *et al.* 2010; Schiopu and Gavrilescu 2010; Singh *et al.* 2010a). Though many of the articles cited in this review have demonstrated the tendency of certain dyes to desorb under specified conditions of pH (for instance, see Acemiglu *et al.* 2010; Yu *et al.* 2009a; Patel and Suresh 2008; Mao *et al.* 2009b), almost none of them have considered what happens at near-neutral pH, *i.e.* at typical pH values of soil. State-of-the-art landfill facilities contain leachates, and even recirculate them, promoting their breakdown (Kumar *et al.* 2011).

### *Composting*

It is well known that various natural processes of biodegradation can be accelerated by use of optimized composting conditions (Singh *et al.* 2010b; Kumar 2011). Lange and Mendez-Sanchez (2010) showed that such an approach can be at least partly successful in decolorizing paper mill effluents that contained azo dyes. Other authors have demonstrated the efficacy of compost-derived micro-organisms for the

biodegradation of synthetic dyes in solution (Anastasi *et al.* 2006; Lopez *et al.* 2006). However, there is evidence that compost prepared from dye-containing wastes from textile operations can be harmful to plant life, if added to soil in sufficient amounts (de Araugo *et al.* 2005).

Using composting for the treatment of dye-loaded cellulosic matter can make especially good sense in terms of some typical characteristics of the material. First, it is well known that a wide range of lignocellulosic materials can be used as the prime energy source, a means of providing permeability to a compost bed, and a means to adjust the ratio of carbon to nitrogen elements in the mixture (Hubbe *et al.* 2010; Kumar 2011). The nitrogen content present in various dyestuffs also can be expected to participate in the biochemical processes, catalyzed by a range of enzymes secreted by decay-promoting organisms. In particular, the presence of dye may be helpful in achieving a suitable C:N ratio that promotes effective composting (Nigam *et al.* 2000). Although some composting practices can be subject to leaching, it is possible to carry out the process in a contained system, such that opportunities for leaching are avoided (Tchobanoglous *et al.* 1993; Dougherty 1998). The amount of material can be substantially reduced by composting (Turovskii and Mathai 2006); so even in cases where the composted material is not judged to be saleable, it is likely to be more suitable or less burdensome for various subsequent treatment steps such as landfilling or incineration (see the previous two subsections).

#### *Wet enzymatic treatment*

One of the drawbacks of composting options, as just outlined, is that the cellulose-based sorbent becomes consumed rather than being restored to usefulness. Most previous studies aimed at regeneration of the biosorbent have employed conditions of pH such that at least certain dyestuffs can be desorbed (see many examples listed in Table A); however, as already discussed, such practices merely release dyes back into solution, rather than destroying them. With this in mind, there is a need to investigate aqueous enzymatic systems in terms of their efficacy in breaking down dyes while they are being held on the solid surfaces. Studies of this type are rare (Knutson and Ragauskas 2004; Rodríguez-Couto *et al.* 2009; Ozmen and Yesilada 2012), yet the approach has shown considerable promise. There is also substantial evidence that laccases can decolorize a wide range of dyes (Anastasi *et al.* 2006; Rodríguez-Couto 2009; Madhavi and Lele 2009; Papinutti and Forchiassin 2010). Rodríguez-Couto *et al.* (2009) demonstrated the feasibility of a two-step process, in which dye from effluent was first sorbed onto biomass, and then the mixture was treated with white-rot fungus, with subsequent in-situ release of laccase, leading to decoloration. A key concern, which will require study, is whether enzymatic treatments can be relied upon to treat systems that contain an unpredictable variety of dyestuffs, such as is likely to be encountered in practice. Also, the issue of contact inhibition, the loss of efficiency of enzymes due to their adsorption onto system components other than the dye to be decolorized, needs to be a concern when considering technologies to treat beds of dye-bearing cellulosic materials. The tendency of laccase to adsorb onto lignocellulosic surfaces is well documented (Saarinen *et al.* 2009; Papinutti and Forchiassin 2010). Though it is possible to modify laccase enzymes so that they will not adsorb to as great an extent onto solid

surfaces (Schoeder *et al.* 2006), so far it appears that such systems have been considered only for the washing of textiles.

Another approach, also employing cellulosic materials, involves immobilization of microbes onto a fixed bed of substrate, through which the contaminated water is passed under controlled conditions (Georgiou and Aivasidis 2005). The cited researchers found that they could achieve good results, even with low residence times, when they controlled the pH by addition of acetic acid, which tends to buffer the pH in a favorable range for the water they were dealing with. It was not clear from the results whether the observed effects were related to adsorption or to molecular degradation.

Conventional wastewater treatment technology, with the use of activated sludge, can be viewed as a way to achieve roughly the same results as enzymatic treatment, but at lower cost. As noted earlier, dyestuffs are often toxic to the micro-organisms that carry out biodegradation, and this is one of the inherent problems with such approaches. In addition, many synthetic dyestuffs have been designed so that they are difficult to degrade (Chakroborty 2010). However, there are many aspects of conventional wastewater treatment that show promise for advanced applications, such as the treatment of effluent from textile facilities. For instance, membranes can be used to prevent the escape of dye molecules from wastewater systems into receiving waters (Lau and Ismail 2009). By such means the dyestuffs can be recirculated back through the system, making it more likely that they will be substantially biodegraded. Substantial progress has been achieved with respect to controlling the fouling of such membranes (Lau and Ismail 2009).

#### *Wet oxidation*

Another kind of approach that could be used to destroy dyes in place, while they are held on the surfaces of a biosorbent in the presence of aqueous solution, is oxidation. In other words, the idea is to oxidatively bleach the bed of biosorbent, breaking down and detoxifying any dye materials that are present. An advantage of this approach is that one can benefit from progress made during many years of development of bleaching technologies (Bajpai 2005; Ramos *et al.* 2008; Suess 2010). It is a common industrial practice to oxidatively bleach left-over dyed paper pulp, *i.e.* “broke”, so that it can be used again in the production of paper products having different colors (Cheek 1991; Walsh *et al.* 1993). Yet other researchers have shown that oxidative bleaching can be used to decolor wastewater from paper mills or textile facilities (Tozer 1993; Wang *et al.* 2006). Electrochemical oxidation of dyes has been investigated in various studies (Carvalho *et al.* 2011). Atchariyawut *et al.* (2009) showed that it is feasible to use a membrane contact system for oxidative decoloration of waters containing reactive dyes. Ozone was found to be an effective decoloring agent under such conditions. Other studies have shown favorable effects when using UV radiation in the presence of catalysts such as zinc oxide (Sil and Chakrabarti 2010). Tozer (1993) noted that one of the advantages of bleaching technologies is that they do not generally give rise to a substantial amount of additional solid waste.

Salazar *et al.* (2011) showed that the effectiveness of dye oxidation in wastewater can be improved by combining a treatment with Fenton’s reagent (ferrous ion with electrochemically generated hydrogen peroxide) and the ultraviolet component of solar



radiation. However, aggressive conditions of treatment, involving relatively high costs, were required in order to break down essentially all of the dye present. One of the key issues in need of research is whether such systems can be carried out in the presence of a biosorbent. For instance, one possibility is to generate the reactive species and then allow the reagent to flow through a packed bed of the biosorbent to be decolorized.

## CLOSING COMMENTS

Based on the reviewed publications it appears that the use of cellulosic materials to remove waste dyes from aqueous systems has considerable promise, but there are some aspects of the overall technology that will require greater attention. There are very large ranges in the capacities of different biosorbent materials to collect different types of dye, so it is still important to search for and develop more effective biosorbents.

Though activated carbon products appear to provide reliable sorption with generally good efficiency, the survey of published information showed that comparable or even superior results often can be achieved with lower-cost byproducts of fermentation, agricultural processing, aquatic plants, *etc.* But the latter materials typically are not as storage-stable and handy to use as activated carbon in its powder form.

There is a need for life-cycle analyses that consider various fates of the biosorbent after it has been used. Since there appears to be little or no value remaining in dilute dyes after they have become mixed into wastewater, there is a need to consider such strategies as oxidative or enzymatic bleaching to regenerate biosorbents that have been used to collect dye materials. In any case, it appears that research related to the biosorption of dyes will continue at a rapid pace, and that there are some priority areas of research that are likely to help facilitate a greater degree of implementation of the technology in the future.

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## APPENDIX

**Table A.** Tabulation of Research Publications for Removal of Dyes from Dilute Aqueous Solution by Use of Raw or Modified Lignocellulosic Materials (See notes below table for key to abbreviations)

Dye Class	Dye	Biomass class	Biomass	Dried, never, heat	Modification	Best pH	Best temperature	Isotherm best fit	Adsorp. capac. (mg/g)	Rate law best fit	Functional groups	Thermodynamics	Key Findings	Author (year)
Ba	BR	Tx	Jute > cotton > viscose	D	Carb-oxyl	High	Low	F	1-5 nat; 25+ mod	–	–	–	Esterification with $\alpha$ -carboxylic acids (citric, malic, tartaric) promoted dye uptake	Abo-Shosha <i>et al.</i> (2002)
Ba	MB	F	<i>Aspergillus</i>	H	–	High	Low	F	3.4	–	–	–	Desorption at pH 3	Acemiglu <i>et al.</i> 2010
Ba	CR	AC	Act. carbon	–	none	Low	Hi	F	40-46	2 <sup>nd</sup>	Yes O	–	Oxygen-containing groups key	Ahmad & Kumar 2010
Ba	MB	AC	Act. carbon	–	Flue gas	–	(30)	L	91	–	–	–	–	Ahmad <i>et al.</i> 2007
Ba	MB	W	Sawdust, meranti	D	–	8-10	Hi	L	120-159	2 <sup>nd</sup>	–	En	Endothermic process	Ahmad <i>et al.</i> 2009a

Dye Class	Dye	Biomass class	Biomass	Dried, never, heat	Modification	Best pH	Best temperature	Isotherm best fit	Adsorp. capac. (mg/g)	Rate law best fit	Functional groups	Thermodynamics	Key Findings	Author (year)
Ba	MB	Ag	<i>Pyracantha</i> berries	H	ground	>6	45	L	128	2 <sup>nd</sup>	Yes	En	Endothermic process; unaffected by salt	Akar <i>et al.</i> 2009d
Ba	MB	F	<i>Rhizopus a.</i>		SDS	10	—	F	370-1667	2 <sup>nd</sup>	—	—	—	Aksu <i>et al.</i> 2010
Ba	MG	Ag	<i>Eriobotrya japon.</i> seed	H	Crush	5	50	L	57	2 <sup>nd</sup>	—	En	—	Aksakal <i>et al.</i> 2009
Ba	MB	N	Date pits	H	Crush	6-8	—	F	278	—	Yes	—	H-bonding & electro-static; particle size had little effect; 2 sites	Al-Gouti <i>et al.</i> 2010
Ba	3 dyes	Pe	Peat	D	Milled	—	(18)	R-P	300-700	—	—	—	Tabulated data	Allen <i>et al.</i> 2004
Ba	3 dyes	Pl	Kudzu	D	8 mm	—	—	—	20	2 <sup>nd</sup>	—	—	10 m <sup>2</sup> /g surface area	Allen <i>et al.</i> 2005
Ba	MG	Ag	<i>Luffa cylindrica</i>	H	NaOH	>5	Hi	L	9-25	2 <sup>nd</sup>	Yes	En	—	Altinisik <i>et al.</i> 2010
Ba	MB	Ag	Banana & orange peel	H	Crush	6-8; >6	—	F, L	12-14	—	—	—	Diffusion limits rates	Annandurai <i>et al.</i> 2002
Ba	RhB	Ag	Banana & orange peel	H	Crush	6-8; >6	—	F, L	7-9	—	—	—	Diffusion limits rates	Annandurai <i>et al.</i> 2002
Ba	MV	Ag	Banana & orange peel	H	Crush	6-8; >6	—	F, L	4-7	—	—	—	Diffusion limits rates	Annandurai <i>et al.</i> 2002
Ba	BY	Al	<i>Caulerpa s.</i> algae	D	Not ground	>4	Low	F	27	2 <sup>nd</sup>	—	Ex	—	Aravindhan <i>et al.</i> 2007

Dye Class	Dye	Biomass class	Biomass	Dried, never, heat	Modification	Best pH	Best temperature	Isotherm best fit	Adsorp. capac. (mg/g)	Rate law best fit	Functional groups	Thermodynamics	Key Findings	Author (year)
Ba	MB, "green"	AC	Act. Carbon	–	ZnCl <sub>2</sub>	–	–	L	50-290	–	O	–	Surface area enhanced, better sorption	Asadullah <i>et al.</i> 2010
Ba	BB41	B	<i>Bacillus macerans</i>	–	–	10	25	L	89	2 <sup>nd</sup>	–	En	–	Atar <i>et al.</i> 2008
Ba	MB	N	Date pits, raw, active.	H	Ground; Carbon	High	Low	L	15-50	2 <sup>nd</sup>	–	Ex	Raw biomass most effective	Banat <i>et al.</i> 2003
Ba	MB	Tx	Jute proc. waste	H	–	High	–	L, F	22	–	–	–	–	Banerjee & Dastidar 2005
Ba	BB41	Ma	<i>Myriophyll. spicatum</i>	–	Dead	–	–	F	–	2 <sup>nd</sup>	–	–	Diffusion	Basibuyuk <i>et al.</i> 2007
Ba	MB, BR22	W	Beech sawdust	H	CaCl <sub>2</sub>	8	(23)	F	10, 20	–	–	–	CaCl <sub>2</sub> treatmt. helped.	Batzias & S 2004
Ba	MB, BR22	W	Beech sawdust	–	Acid hydrol.	>8	(23)	F	1.6-3.1	1 <sup>st</sup>	–	–	Hemicellulose removal by acid, more uptake	Batzias & S 2007a
Ba	MB	W	Beech sawdust	–	–	High	(23)	F	–	–	–	–	PZC theory	Batzias & S 2007b
Ba	MB	W	Beech sawdust	–	–	–	(23)	F	5-10	–	–	–	Salt treatment greatly increased dye uptake.	Batzias & S 2007c
Ba	MG	Ma	<i>Caulerpa racem.</i> alga	H	Ground	–	45	F	17	2 <sup>nd</sup>	–	–	Physisorption	Bekci <i>et al.</i> 2009

Dye Class	Dye	Biomass class	Biomass	Dried, never, heat	Modification	Best pH	Best temperature	Isotherm best fit	Adsorp. capac. (mg/g)	Rate law best fit	Functional groups	Thermodynamics	Key Findings	Author (year)
Ba	MB	W	<i>Azadirachta indica</i> neem	H	Crush	No effect	high	L, F	5-23	1 <sup>st</sup>	–	En	Simultaneous surface & pore adsorption	Bhattacharyya & Sharma 2005
Ba	MV, fuchsin	F	Fungal mix	H	Crush	Weak effect	–	–	6-25	–	–	–	HCl desorption	Bhole <i>et al.</i> 2004
Ba	CR	F	<i>Trametes versicolor</i>	A	Pellets	Low	(30)	L	52	2 <sup>nd</sup>	–	–	Autoclaving helped; acid & alkaline did not.	Binupriya <i>et al.</i> 2008b
Ba	MB	Ag	Wheat shells	H	ground	>4	50		16-22	–	–	En	–	Bulut & Aydn 2006
Ba	MB	AC	Hickory husk	–	ZnCl <sub>2</sub>	–	–	–	400	–	–	–	Optimized activation	Cao <i>et al.</i> 2010
Ba	MB	Ma	<i>Caulerpa racemosa</i>	H	ground	high	(27)	L	5	2 <sup>nd</sup>	–	–	Chemisorption; little effect of temperature	Cengiz & Cavas 2008
Ba	MV	Ma	<i>Posidonia oceanica</i>	–	–	No effect	45	L	119	2 <sup>nd</sup>	–	En	–	Cengiz & Cavas 2010
Ba	BB41	Ma	<i>Posidonia oceanica</i>	H	Carboxy-methyl	>6	30	L	205-3200	1 <sup>st</sup>	–	Ex	Carboxymeth. helped.	Chadlia & Farouk 2007
Ba	MB	AC	Durian shell	–	–	–	(30)	–	289	–	–	–	–	Chandra <i>et al.</i> 2007
Ba	MG	SI	Anaerobic sludge	N, A	washed	>5	Low	L, F	60	2 <sup>nd</sup>	–	Ex	Salt had negative effect	Cheng <i>et al.</i> 2008
Ba	MB	AC	Ash from rice husk	–	As is	>6	(30)	RP	181	–	–	–	–	Chowdhury <i>et al.</i> 2009

Dye Class	Dye	Biomass class	Biomass	Dried, never, heat	Modification	Best pH	Best temperature	Isotherm best fit	Adsorp. capac. (mg/g)	Rate law best fit	Functional groups	Thermodynamics	Key Findings	Author (year)
Ba	MG	Al	<i>Cormarium</i> sp.	L	—	>5	high	—	8	—	—	—	—	Daneshvar <i>et al.</i> 2007
Ba	RhB	F	<i>Rhizopus oryzae</i>	L, A	Starved, etc.	>7	40	L	40	—	—	—	No biodegradation	Das <i>et al.</i> 2006
Ba	Astrazon Y7GL	AC	Apricot stone AC	—	—	10	50	—	221	—	—	—	—	Demirbas <i>et al.</i> 2008
Ba	MB	AC	Cotton stalk AC	—	ZnCl <sub>2</sub>	9	(25)	L	193	—	—	—	—	Deng <i>et al.</i> 2009
Ba	BR46	W	<i>Pinus brutia</i> Ten	—	—	8	(45)	—	74	—	—	—	—	Deniz <i>et al.</i> 2011
Ba	MG	F	<i>Penicillium</i>	—	—	6	20	L	—	—	—	—	—	Diaconu <i>et al.</i> 2010
Ba	MB	N	Hazelnut shell	—	crushed	high	high	L	68-74	2 <sup>nd</sup>	—	En	Salt helped	Dogan <i>et al.</i> 2008
Ba	MB	N	Hazelnut shell	—	crushed	high	high	—	100	2 <sup>nd</sup>	—	En	Diffusion control	Dogan <i>et al.</i> 2009
Ba	MB	Al	<i>Ulothrix</i> algae	H	—	>6	Low	—	30-87	2 <sup>nd</sup>	—	Ex	—	Dogar <i>et al.</i> 2010
Ba	BB86	W	Oak sawdust	—	As is	8	(25)	L	33	1 <sup>st</sup>	—	—	—	Dullman <i>et al.</i> 2002
Ba	BB86	W	Beech sawdust	—	Milled	5-7.5	—	L	137	2 <sup>nd</sup>	—	—	—	Dulman & C 2009
Ba	MB	Ma	<i>Posidonia oceanica</i>	—	—	6.5	(25)	—	286	—	—	—	—	Dural <i>et al.</i> 2011

Dye Class	Dye	Biomass class	Biomass	Dried, never, heat	Modification	Best pH	Best temperature	Isotherm best fit	Adsorp. capac. (mg/g)	Rate law best fit	Functional groups	Thermodynamics	Key Findings	Author (year)
Ba	MB	Ma	Water hyacinth	D	HNO <sub>3</sub>	–	No eff.	–	350	2 <sup>nd</sup>	–	–	Sun dried; diffusion control	El-Khaiary 2007
Ba	MB	W	Oak sawdust	H	NaOH, H <sub>2</sub> SO <sub>4</sub>	8	(25)	L	87-93	2 <sup>nd</sup>	–	En	Intra-particle diffusion	El-Latif & I. 2009
Ba	Astraz B	F, AC	Yeast; Act. C	H	ground	high	high	L	70, 18	–	–	En	–	Farah <i>et al.</i> 2007
Ba	BR14	AC	HNO <sub>3</sub> oxidation	–	–	–	(25)	–	550-710	–	Acid & basic	–	Not just electrostatic	Faria <i>et al.</i> 2004
Ba	MB	N	<i>Sesamum indicum</i> L.	H	Crush	3.5-10.5	–	L	360	2 <sup>nd</sup>	several	–	–	Feng <i>et al.</i> 2011
Ba	MB	N, W	Hazelnut sh Sawdusts	H	ground	–	(20)	L	25-76	2 <sup>nd</sup>	–	–	–	Ferrero 2007
Ba	MB	AC	Act. C from oil palm	–	Micro-wave	as-is	(30)	–	312	–	–	–	–	Foo & Hameed 2011
Ba	MG (BG4)	AC	Coffee press cake	–	Micro-wave	>5	(25)	L	197-208	2 <sup>nd</sup>	–	–	–	Franca <i>et al.</i> 2010
Ba	BB9	F	<i>Aspergillus niger</i>	A	–	>5	–	L	19	–	–	–	Autoclaving helped sorp.	Fu & Viraraghavan 2000
Ba	BB9	F	<i>Aspergillus niger</i>	A	ground	(6)	–	–	0.4-16	–	–	–	Functional groups control adsorption	Fu & Viraraghavan 2002a
Ba	RhB	AC	Act. C from bagasse	–	H <sub>3</sub> PO <sub>4</sub> or KOH	6	20	L, F	199; 22	2 <sup>nd</sup>	–	En	Acid activation much better; chemisorption	Gad & El-Sayed 2009

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Ba	MG	W, AC	<i>Dalbergia sissoo</i> dust	H	CH <sub>2</sub> O, H <sub>2</sub> SO <sub>4</sub>	6-9	(26)	—	—	1 <sup>st</sup>	—	—	—	Garg <i>et al.</i> 2003
Ba	MG	W, AC	<i>Prosopis cineraria</i> sawdust; Act. carbon	D	CH <sub>2</sub> O, H <sub>2</sub> SO <sub>4</sub>	6-10	(27)	—	25-55	—	—	—	—	Garg <i>et al.</i> 2004
Ba	—	AC	Act. C from <i>E. rigida</i>	—	H <sub>2</sub> SO <sub>4</sub> activat.	6-10	40	L	114	2 <sup>nd</sup>	—	—	Diffusion controlled	Gercel <i>et al.</i> 2007
Ba	—	Ag	Rice straw	H	Citric ac ester	>4	(20)	L, F	94, 256	1 <sup>st</sup>	—	—	Citric acid treatment increased uptake ~3X.	Gong <i>et al.</i> 2006
Ba	MB, BCB, NR	N	Peanut hull	H	Crush, citric ac ester	(5)	~20	—	—	—	—	—	Citric acid treat helped Ba dye, not anionics; methylation: no effect.	Gong <i>et al.</i> 2005
Ba	MB	Ag	Kohlrabi peel	—	Granular	5	(20)	—	112	—	—	—	—	Gong <i>et al.</i> 2007
Ba	AO, MG	Ag	Soybean hull	H	Esteri-fied	>6	low	L	238, 179	2 <sup>nd</sup>	—	Ex	—	Gong <i>et al.</i> 2008
Ba	Fuchsin	Ash	Bottom ash, power plant	—	—	>4	50	L	2-3	2 <sup>nd</sup>	—	En	—	Gupta <i>et al.</i> 2008
Ba	MB	W	Cedar sawdust	—	—	7	(20)	L	142	2 <sup>nd</sup>	—	Ex	Diffusion control.	Hamdaoui 2005
Ba	MB	Ag	Papaya seed	H	Crush	4-10	(30)	L	556	2 <sup>nd</sup>	—	—	—	Hameed 2009a

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Ba	MB	Ag	Jackfruit peel	H	Ground	>3	(30)	L	286	2 <sup>nd</sup>	–	–	–	Hameed 2009b
Ba	MB	Ag	Garlic peel	H	Crush	4-12	(30)	F	83-143	2 <sup>nd</sup>	–	En	–	Hameed & Ahmad 2009
Ba	MB	AC	Act C from rattan sawd	–	KOH	–	(30)	L	294	2 <sup>nd</sup>	–	–	–	Hameed <i>et al.</i> 2007b
Ba	BB3	AC	Act C from <i>Hevea</i> seed	–	KOH	–	(30)	F	227	2 <sup>nd</sup>	–	–	–	Hameed & Daud 2008
Ba	MB	AC	Act. C from bamboo	–	KOH, CO <sub>2</sub>	(7)	(30)	L	454	2 <sup>nd</sup>	–	–	–	Hameed <i>et al.</i> 2007c
Ba	MG	AC	Act. C from bamboo	–	K <sub>2</sub> OC <sub>3</sub> + CO <sub>2</sub>	3-8	(30)	L	264	2 <sup>nd</sup>	–	–	Pore diffusion; week pH effect	Hameed & El-K. 2008a
Ba	MG	W	Rattan sawdust	H	Crushed	10-12	(30)	L	63	1 <sup>st</sup>	–	–	Film & pore diffusion	Hameed & El-K. 2008b
Ba	MB	Ag	<i>Citrus gran-dis</i> peel	H	Ground	8	(30)	L	345	2 <sup>nd</sup>	–	–	–	Hameed <i>et al.</i> 2008a
Ba	MB	W	Bamboo waste	–	Nitric acid	–	–	L	88	2 <sup>nd</sup>	–	–	Intraparticle diffusion	Hameed & Tan 2010
Ba	MB	W	Banana stalk	H	Ground	4-12	(30)	L	244	2 <sup>nd</sup>	–	–	–	Hameed <i>et al.</i> 2008b
Ba	MB	Ag	<i>Cocos nucifera</i>	H	Ground	>10	(30)	L	91	2 <sup>nd</sup>	–	–	–	Hameed <i>et al.</i> 2008c
Ba	MB	Ag	Rice husk	H	–	–	–	–	4	–	–	–	Bed breakthrough predictions.	Han <i>et al.</i> 2007a
Ba	BM	PI	Phoenix tree leaf	H	Ground	4.5-10	50	L	81-90	–	–	En	Salt adversely affected uptake.	Han et al. 2007b



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Ba	BY21	Ag	<i>Linum usitatissim.</i>	–	–	–	–	T	77	2 <sup>nd</sup>	–	–	–	Hassanein & K 2010
Ba	MG	AC	AC	–	–	5-6	any	L	10	–	–	nul	Ca <sup>2+</sup> interferes; desorp by acid.	Hema & Arivoli 2008
Ba	RhB	Ma	<i>Turbinaria c</i> seaweed	H	Powder vs. not	3-7	high	L	80	–	–	En	Powder sorbed more; dye not cat if pH>4.2	Hii <i>et al.</i> 2009
Ba	BR13	PI	Tree fern	H	Crushed	(5)	high	L	408	–	–	En	Smaller particles sorbed more.	Ho <i>et al.</i> 2005a
Ba	BV10, BV1, BG4	Ag	Sugarcane dust	H	Ground	–	(25)	L, RP	50, 14, 21	–	–	–	–	Ho <i>et al.</i> 2005b
Ba	BR2	PI	Pith from bagasse	–	Crushed	–	–	–	83	2 <sup>nd</sup>	–	–	Important early work with rate law.	Ho & McKay 1999
Ba	CR	PI	Cattail root	H	ground	(7)	20	L	31-39	2 <sup>nd</sup>	–	Ex	Salt aided sorption	Hu <i>et al.</i> 2010
Ba	MB	Ag	Corn stalk	–	Carbon-ized	–	–	–	4-5	–	–	–	Carbonation increased sorption by ca. 20%	Husseien <i>et al.</i> 2009
Ba	MB others	AC	Act. charcoal	–	Fluka	low	low	L	18-24	–	–	Ex	pH effects depend on dye type	Iqbal & Ashiq 2007

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Ba	MG	AC	Act. charcoal	–	Fluka	10	low	L	18-24	–	–	Ex	pH effects depend on dye type	Iqbal & Ashiq 2007
Ba	MB	F	<i>Pleurotus ostreatus</i>	–	–	11	–	L	70	–	–	–	–	Isek <i>et al.</i> 2008
Ba	MB, CV	N	<i>Feronia a</i> shell	H	ground	high	(32)	L	130, 95	2 <sup>nd</sup>	O atoms	–	–	Jain & Jayaram 2010
Ba	MB	W	<i>Picea</i> wood shavings	H	HCl, CO <sub>3</sub> Na <sub>2</sub> HPO <sub>4</sub>	high	–	L-F	7-47	–	–	–	Alkaline treatments helped MB sorption.	Janoš <i>et al.</i> 2009
Ba	BB41	AC	Pyrolysis char sludge	–	N <sub>2</sub> or CO <sub>2</sub>	(6-7)	–	L	200-558	–	–	–	High-temp treatment increased sorption	Jindarom <i>et al.</i> 2007
Ba	BR22	AC	Act. C from plum kernel	–	Steam	–	(30)	–	150-500	2 <sup>nd</sup>	–	–	Higher temp of prep helped; diffusion control	Juang <i>et al.</i> 2000
Ba	MB	AC	Various	–	Acid-treated	high	(30)	L, F	165-980	1 <sup>st</sup>	–	–	–	Kannan & S 2001
Ba	MB	AC	Sunflower oil cake	–	H <sub>2</sub> SO <sub>4</sub>	>6	25	L	10-16	2 <sup>nd</sup>	–	–	–	Karagoz & Tay 2008
Ba	MB	AC	Coir pith carbon	–	Not activated	7	(35)	L	6	2 <sup>nd</sup>	–	–	Diffusion	Kavitha & N 2007b

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Ba	BR2 safranin	F	<i>Aspergillus wentii</i>	–	–	–	–	L	–	–	–	–	Monolayer sorption, multi-parameter fits	Khambhaty <i>et al.</i> 2011
Ba	MG	W	Neem sawdust	–	–	7	(30)		4	–	–	–	–	Khatti & Singh 2009
Ba	MB	AC	Act. C pow. fibers	–	Steam activat.	–	–	L	225, 480	–	–	–	Bottle-neck model	Ko <i>et al.</i> 2002
Ba	MB	AC	Lignin (lignite)	–	–	11		L	–	2 <sup>nd</sup>	–	–	–	Kriaa <i>et al.</i> 2011
Ba	MB	PI	<i>Paspalum notatum</i>	D	ground	8 (high)	(30)	L	31	2 <sup>nd</sup>	–	–	–	Kumar & Porkodi 2007
Ba	MG	AC	Act. C	–	Merck	8	(32)	L, R-P	480	2 <sup>nd</sup>	–	–	Pseudo-isotherms, nonlinear fits	Kumar & Sivanesan 2006
Ba	MG	W	<i>Hevea brasiliensis</i>	H	sawdust	–	(32)	L, R-P	33	–	–	–	Nonlinear fits	Kumar & Sivanesan 2007
Ba	MG	Al	<i>Pithophora sp.</i> (algae)	D	milled; 300 °C	>5	–	L	12-118	2 <sup>nd</sup>	–	–	Thermal activation helped sorption.	Kumar <i>et al.</i> 2005
Ba	MG	Al	<i>Pithophora sp.</i> (algae)	D	Milled	6	(30)	F	65	1 <sup>st</sup>	–	–	Diffusion control	Kumar <i>et al.</i> 2006
Ba	MB	Ag	Leaves, husks, etc.	H	Crush	–	–	L	100-400	2 <sup>nd</sup>	–	–	Divalent ion competition	Kumar <i>et al.</i> 2010
Ba	BR46, BY28	W	HW vs. SW sawdust	H	Sawdust	–	(25)	L	20-33	1 <sup>st</sup>	–	–	Filtration-adsorption	Laasri <i>et al.</i> 2007
Ba	RhB	AC	<i>Parthenium</i>	H	H <sub>2</sub> SO <sub>4</sub>	(7)	(22)	L	17-25	2 <sup>nd</sup>	FTIR	–	–	Lata <i>et al.</i> 2008a

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Ba	MG	AC	<i>Parthenium</i>	H	H <sub>2</sub> SO <sub>4</sub>	3-10	(22)	L, F	19	2 <sup>nd</sup>	-	-	-	Lata <i>et al.</i> 2008b
Ba	RhB	PI, AC	<i>Parthenium</i> & its act. C	D	CH <sub>2</sub> O; H <sub>3</sub> PO <sub>4</sub>	(7)	(36)	L, F	29 59	2 <sup>nd</sup>	Yes	-	Not sensitive to pH	Lata <i>et al.</i> 2008c
Ba	MV	PI	Tea powder	-	-	-	(25)	-	-	2 <sup>nd</sup>	-	-	-	Li <i>et al.</i> 2010
Ba	BB9	AC	Act. C	-	KOH act, CO <sub>2</sub>	-	(26)	-	200-400	2 <sup>nd</sup>	-	-	Diffusion control	Ling <i>et al.</i> 2011
Ba	MB, VB	W	<i>Eichhornia crassipes</i>	-	-	>6	-	L	129, 145	1st	-	-	Water hyacinth root.	Low <i>et al.</i> 1995
Ba	MB	AC	Act. C from coir dust	-	ZnCl <sub>2</sub> 800 °C	4-8	(25)	-	13-14	2 <sup>nd</sup>	Acid & base	-	Diffusion control	Macedo <i>et al.</i> 2006
Ba	CV	Ag AC	Sugarcane baggase	H	CH <sub>2</sub> O, H <sub>2</sub> SO <sub>4</sub>	(7)	30, 60	F	-	-	-	Ex, En	Carbonized at low heat	Mahesh <i>et al.</i> 2010
Ba	MB, CV	AC	Appler rind	-	-	high	-	-	-	-	-	-	-	Malarvizhi & Ho 2010
Ba	MG	AC	Bagasse fly ash & AC	-	-	7	(30)	F, R-P	170, 8-42	2 <sup>nd</sup>	-	-	-	Mall <i>et al.</i> 2005
Ba	MV (BV1)	FA	Fly ash fr. bagasse	-	Sieving	9	(30)	R-P	26	2 <sup>nd</sup>	-	-	-	Mall <i>et al.</i> 2006a
Ba	BY2, CR, MV	AC	Fertilizer plant waste	-	Sieving	7	(25)	R-P	65, 25, 46	2 <sup>nd</sup>	-	-	Regeneration by pH did not work.	Mall <i>et al.</i> 2006b
Ba	BG4	AC	Rice husk ash	-	-	3	45	L, R-P	25	2 <sup>nd</sup>	-	En	-	Mane <i>et al.</i> 2007a
Ba	BB3	B	<i>Corynebact glutamicum</i>	H	Powder	10	(25)	L	28-78	-	-	-	From MSG ferment.; can desorb, low pH.	Mao <i>et al.</i> 2008

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Ba	BB3	B	<i>Corynebact glutamicum</i>	H	Citric acid	>7	(25)	R-P	41-52	1 <sup>st</sup> 2 <sup>nd</sup>	–	–	–	Mao <i>et al.</i> 2010
Ba	MB	B	<i>Corynebact glutamicum</i>	–	–	–	–	L	–	–	COO <sup>-</sup>	–	–	Mao <i>et al.</i> 2009c
Ba	BB159 + BB3	Al	<i>Caulerpa lentillifera</i>	H	–	–	51	L, F	31-49	2 <sup>nd</sup>	–	–	–	Marungrueng & P 2006
Ba	3 dyes MB	Al	<i>Caulerpa lentillifera</i>	H	–	(7)	(25)	L	39-48, 417	2 <sup>nd</sup>	–	–	Better than activated C	Marungrueng & P 2007
Ba	Alcian B	F	<i>Fomitopsis carnea</i> fung	–	ground	high	40	L	11-24	–	–	En	Salt hurt uptake; acid desorption.	Maurya & Mittal 2009
Ba	Saffra., MB	Ag	Bark, rice husk, cotton	H	–	–	(30)	–	10-70, 20-85	–	–	–	Bark > cotton > rice husk	McKay <i>et al.</i> 1986
Ba	–	AC	Granular	–	–	–	(20)	L, F	159-309	–	–	–	Diffusion	Meshko <i>et al.</i> 2001
Ba	RhB	AC	Palm shell	H	NaOH post	3	50	L	1-2	2 <sup>nd</sup>	–	–	–	Mohammadi <i>et al.</i> 2010
Ba	MB	AC	Char from bamboo	–	Pre-crushed	–	–	RP	320	–	–	–	Not very effective for two acid dyes	Mui <i>et al.</i> 2010
Ba	MB	B	<i>Streptomyces rimosus</i>	H	NaOCl oxidized	–	20	F,L	24-34	2 <sup>nd</sup>	–	Ex	–	Nacera & Aicha 2006
Ba	RhB	Ag	Orange peel	D	Crush	Low	(29)	L, F	3	1 <sup>st</sup>	–	–	Desorption at alkaline pH	Namasivayam <i>et al.</i> 1996
Ba	RhB, MB	Ag	Coir pith	D	Ground	3, >5	(30)	L, F	203	1 <sup>st</sup>	–	–	–	Namasivayam <i>et al.</i> 2001

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Ba	CR	AC	Act. C from coir pith	D	Pre-grnd, ZnCl <sub>2</sub>	Low	60	L, F	7	2 <sup>nd</sup>	—	En	Weak temperature dependence	Namasivaya m & K 2002
Ba	CR	AC	Act. C from coir pith	D	Pre-grnd, ZnCl <sub>2</sub>	—	(35)	—	—	—	—	—	Basic, reactive, acid dyes all sorbed.	Namasivaya m & S 2006
Ba	CR	SI	Biogas waste, cow	D	ground	2.3-9.4	(30)	F	5-20	1 <sup>st</sup>	—	—	Lower pH better	Namasivaya m & Y 1992
Ba	BR18, BY11	AC	Act. C fiber	—	—	—	(30)	L, F	90, 120	—	COO <sup>-</sup>	—	Alginate groups	Nasr <i>et al.</i> 2006
Ba	R11055 Astraz.	Ag	Bagasse pith, cobb	D	—	—	(23-27)	L	76-160, 94-167	—	—	—	—	Nassar & E 1991
Ba	MB	Ag	Rejcted tea	—	—	(4)	(30)	—	147	—	—	—	—	Nasuha <i>et al.</i> 2010
Ba	MB	Al	<i>Enteromor.</i> alga	H	—	6-10	(30)	L, RP	274	—	—	—	—	Ncibi <i>et al.</i> 2009a
Ba	MB	Ma	<i>Posidonia oceanica</i>	H	—	6-9	(30)	L	5	2 <sup>nd</sup>	—	—	—	Ncibi <i>et al.</i> 2007a
Ba	MG	AC	Act. C from <i>Borassus a</i>	—	H <sub>2</sub> SO <sub>4</sub>	6-8	40	L	5-50	2 <sup>nd</sup>	—	En	—	Nethaji <i>et al.</i> 2010
Ba	MB, MV	W	<i>Mansonía a</i> sawdust	H	ground	>10	(26)	—	15-30	2 <sup>nd</sup>	—	—	Mass transfer and interparticle diffusion	Ofomaja 2008

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Ba	MV	W	<i>Mansonia a</i> sawdust	H	ground	high	56	L, RP	15-25	–	–	En	At pH above iep of substrate	Ofomaja & Ho 2008
Ba	MB	Ag	Coffee husk	H	–	any	–	L, F	73-111	–	–	Nul	Not sensitive to temperature	Oliveira <i>et al.</i> 2008
Ba	MB, MG, CV	AC	Activated C from apricot	–	ZnCl <sub>2</sub>	–	40	–	79-90, 89-112, 53-64	2 <sup>nd</sup>	–	En	External mass transfer and interparticle diffusion	Onal 2006
Ba	MG	AC	Activated C from lignite	–	KOH	–	50	L	100	2 <sup>nd</sup>	–	En	Mass transfer and interparticle diffusion	Onal <i>et al.</i> 2007
Ba	MG	W	<i>Firmiana simplex</i>	D	Ground	high	(25)	F	1	2 <sup>nd</sup>	–	En	–	Pan & Zhang 2009
Ba	CR, RhB	Ag	Jute stick powder	H	Ground	7	Any	L, F	36, 88	2 <sup>nd</sup>	–	Nul	Interparticle diffusion	Panda <i>et al.</i> 2009
Ba	MG, others	F	<i>Pleurotus ostreatus</i>	N	Laccase produced	3	(30)	–	–	–	–	–	Ezymatic breakdown & sorption important	Papinutti & F 2010
Ba	CV, RhB	Ag	Coir, cane, sawdust	D	Ground	–	(27)	L, F, R-P	10-66, 16-56	2 <sup>nd</sup>	–	–	–	Parab <i>et al.</i> 2009
Ba	MB	Ag	Passion frt; Mandardin	H	Ground	9, 11	25	–	–	–	–	–	Response surface	Pavan <i>et al.</i> 2007
Ba	MB	Ag	Passion frt; Mandardin	H	Ground	high	25	Sips	45	2 <sup>nd</sup>	–	–	Ion exchange	Pavan <i>et al.</i> 2008

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Ba	MB, CV	W	Spruce TMP	N	KMnO <sub>4</sub> oxidized	(7.7)	(20)	–	–	–	–	–	Sorption correlated with COOH groups.	Peterlin <i>et al.</i> 2009a
Ba	MB	Ag	<i>Psidium guajava</i>	D	Ground	7.5	30	L	295	2 <sup>nd</sup>	–	–	–	Ponnusami <i>et al.</i> 2008
Ba	MG, CV	AC	Activated C from jute fib	–	H <sub>3</sub> PO <sub>4</sub>	10	30	L, F	137, 28	2 <sup>nd</sup>	–	–	Surface & pore diffusion	Porkodi & Kumar 2007
Ba	Safra-nin	AC	Activated C fr. corncob	–	H <sub>2</sub> SO <sub>4</sub> , ground	5-9	(30)	L, F	1400	1 <sup>st</sup>	–	En	Smaller particles sorbed more.	Preethi <i>et al.</i> 2006
Ba	MG	AC	Rice husk	–	H <sub>3</sub> PO <sub>4</sub> NaOH	(5-6))	–	L, F; L	56-92	–	–	–	–	Rahman <i>et al.</i> 2005
Ba	–	AC	Activated C fr. cassava	–	H <sub>3</sub> PO <sub>4</sub>	(6-7)	(30)	–	–	–	–	–	Basic dyes removed effectively.	Rajeshkanna <i>n et al.</i> 2001
Ba	MB	Ma	<i>Sargassum muticum</i>	H	Ground, CaCl <sub>2</sub> , HCl	(5.5)	(25)	L	- 237, 279	1 <sup>st</sup>	–	–	–	Rubin <i>et al.</i> 2005
Ba	MR	–	Sugarcane bagasse	–	H <sub>3</sub> PO <sub>4</sub>	3-6	–	–	–	–	–	–	–	Saad <i>et al.</i> 2010
Ba	Rh	F	<i>Trichoderm. harzianum</i>	N	–	8	(30)	L,F	3	2 <sup>nd</sup>	–	–	120 min to equilib., NaOH to regenerate	Sadhasivam <i>et al.</i> 2007a
Ba	MB	F	<i>Trichoderm. viride</i> , loofa	A	Immobil. In loofa	>6	(30)	L	155, 202	2 <sup>nd</sup>	Variou s FTIR	–	Immobilization in loofa helped 30%; HCl regeneration	Saeed <i>et al.</i> 2009



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Ba	Aniline blue, +	F	<i>Saccharom cerevisiae</i>	N, A	Magnet mod.	–	(25)	–	12-228	–	–	–	Various dyes	Safarikova <i>et al.</i> 2005
Ba	CR	Ce	Chomatog. paper	–	–	–	55	L	14-17	2 <sup>nd</sup>	–	En	Intraparticle diffusion limiting	Samiey and Dargahi 2010
Ba	Procion blue	F	<i>Panus fulvus</i>	A	–	2	35	L	40-55	1 <sup>st</sup>	Amine s	–	Acetone desorp, high pH	Sathishkumar <i>et al.</i> 2007
Ba	MG	Ce	Cellulose powder	–	–	7-98	(25)	L, F	2.4	2 <sup>nd</sup>	–	Ex	Regen. with PEO	Sekhar <i>et al.</i> 2009
Ba	CV	AC	Activated carbons	–	H <sub>3</sub> PO <sub>4</sub>	High	(25)	L	60, 86	2 <sup>nd</sup>	–	En	–	Senthikumaa <i>r et al.</i> 2006b
Ba	MB	AC	Activated carbon jute	–	H <sub>3</sub> PO <sub>4</sub>	5-10	(28)	L	226	2 <sup>nd</sup>	–	–	Intraparticle diffusion	Senthikumaa <i>r et al.</i> 2005
Ba	MB	AC	Coconut coir	–	ZnCl <sub>2</sub>	6-8	50	L, etc.	16-21	1 <sup>st</sup>	–	En	–	Sharma & U. 2009
Ba	MG	AC	Rice husk AC	–	–	6	(30)	–	64	–	–	–	–	Sharma <i>et al.</i> 2009
Ba	MG, MB	W	Eucalyptus bark	H	Ground	Any, >6	40	L	298-425 125-156	2 <sup>nd</sup>	–	En	–	Srivastafa & R. 2009
Ba	Astrazon Y7GL	Ag	Wheat bran	–	–	6	(30)	–	69	–	–	–	–	Sulak <i>et al.</i> 2007
Ba	MB	Al	<i>Ulva lactuc. Sargassum</i>	H	Ground	5-9	(25)	L	93	–	–	Ex	–	Tahir <i>et al.</i> 2008
Ba	MB	AC	Activated carbon	–	HCl	(6.5)	30	L	278	2 <sup>nd</sup>	–	En	–	Tan <i>et al.</i> 2007

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Ba	MB	AC	Activated carbon	–	HCl	high	50	L	303	2 <sup>nd</sup>	–	En	–	Tan <i>et al.</i> 2008a
Ba	BO	PI	<i>Azolla filiculoides</i>	H	Ground	4-10	–	L	833	–	–	En	–	Tan <i>et al.</i> 2010
Ba	BO	PI	<i>Azolla filiculoides</i>	H	Ground	7	(30)	L	833	2 <sup>nd</sup>	Amine, COO, +	En	Chemisorption	Tan <i>et al.</i> 2011
Ba	MB	AC	Bagasse, straw	–	H <sub>3</sub> PO <sub>4</sub> NNO <sub>3</sub>	wide	–	–	–	–	–	–	–	Tandon & Nandini 2011
Ba	MB	Ag	<i>Cucumis sativa</i> peel	–	H <sub>2</sub> SO <sub>4</sub> crushed	>6	(27)	L	47	2 <sup>nd</sup>	–	–	–	Thirumalisamy & S. 2010
Ba	MV	F	Bakers' yeast	H	Crushed, Fe <sub>3</sub> O <sub>4</sub> + glutarald.	6	55	L	61	2 <sup>nd</sup>	COO, etc	En	Magnetic collection separation; HOAc regeneration	Tian <i>et al.</i> 2010
Ba	MG	Al	<i>Chlorella</i> algae	D	–	High	(25)	–	9	2 <sup>nd</sup>	–	Nul	Slight effect of agitation; weak pH effect	Tsai & Chen 2010
Ba	MB	AC	Activated C from wood	–	H <sub>3</sub> PO <sub>4</sub>	High	(21)	L	454-625	–	–	–	Loss of functional groups on heating	Tsang <i>et al.</i> 2007
Ba	MB	Ag	Rice husk	H	Ground	High	(32)	L	41	2 <sup>nd</sup>	–	–	Surface & pore diffusion control	Vadivelan & V. K. 2005

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Ba	TB	W	TMP, kraft pulp fibers	N	—	8	—	L	8-27	—	—	—	Adsorption: charge + hydrophobic	Van de Ven <i>et al.</i> 2007
Ba	MB	FA	Fly ash	H	—	>7	(30)	L	6	2 <sup>nd</sup>	—	—	Surface & pore diffusion control	Vasanth K. <i>et al.</i> 2005
Ba	MB	B	<i>Corynebact glutamicum</i>	H	Immobil. p-sulfone	>7	(25)	L	80-190	2 <sup>nd</sup>	—	—	Intraparticle diffusion; Acid regeneration	Vijayaraghavan <i>et al.</i> 2008b
Ba	MB	Al	<i>Gelidium</i> algae	—	Crushed Immobil.: PAN	(6)	(20)	L	74-171	1 <sup>st</sup> 2 <sup>nd</sup>	—	—	—	Vilar <i>et al.</i> 2007
Ba	MB	Al	Marine alga, Macrophyte	—	—	—	(20)	F, L	155, 150	—	—	—	—	Wang <i>et al.</i> 2008a
Ba	MG, MB	Ag	Rice bran, Wheat bran	H	Ground	>4	(20)	L, F	38-69, 16-55	2 <sup>nd</sup>	—	—	Smaller sorbed more.	Wang <i>et al.</i> 2008b
Ba	MG RhB	AC	<i>P. orientale</i> Linn	—	—	5	(25)	—	480 556	—	—	—	—	Wang <i>et al.</i> 2010
Ba	MB, CV, RhB	AC	Activated C	H	HNO <sub>3</sub> HCl	10	50	—	260	2 <sup>nd</sup>	—	En	Nitric treat reduced MB sorption, pKa lowered greatly.	Wang & Zhu 2007
Ba	MB	Ma	<i>Spirodela polyrrhiza</i>	H	—	3-11	(25)	L	112-130	1 <sup>st</sup>	—	—	Intraparticle diffusion	Waranusant. <i>et al.</i> 2003

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Ba	BB3	B	<i>Corynebact glutamicum</i>	H	Poly-amic grafting	Alk.	(25)	L	53-174	–	–	–	Grafting helped greatly.	Won <i>et al.</i> 2009a
Ba	MB	AC	Fir wood char	–	KOH, CO <sub>2</sub>	–	(30)	L	450-650	–	–	–	Elovich rate, low oxide favorable	Wu & Tseng 2006
Ba	MB, BB1	AC	Fir wood char	–	NaOH	–	(30)	L	450-1200	–	–	–	Surface coverage	Wu & Tseng 2008
Ba	MB	AC	Activated C fr. corncob	–	KOH	–	–	L	500-590	–	–	–	Elovich rate	Wu <i>et al.</i> 2011
Ba	BV, BG	F	Brewer's yeast	H	–	>4	30	L	115, 142	2 <sup>nd</sup>	–	Ex	–	Wu <i>et al.</i> 2012
Ba	BY2 BG4		<i>Phaneroch. chrysosp.</i>	–, A	–	4, 4		F	23, 26	–	–	–	–	Yalcin <i>et al.</i> 2010
Ba	MB, RhB	F	Baker's yeast	–	TiO <sub>2</sub> for photo-degrad.	(6)	–	L	50, 25	2 <sup>nd</sup>	–	–	Regeneration by acid; photocatal. degradation	Yu <i>et al.</i> 2009a
Ba	MB, RhB	F	Baker's yeast	–	PMAA	(6.5)	(25)	L	870, 2670	–	–	En	More COOHs: > 10X sorption	Yu <i>et al.</i> 2009b
Ba	Neutral Red	Ag	Corncob	H	Crush	4-7	20-40	L	139-143	2 <sup>nd</sup>	–	–	–	Yu <i>et al.</i> 2011
Ba	MB	AC	Activated C, biogas	–	H <sub>3</sub> PO <sub>4</sub>	(6.5)	(25)	L	345	1 <sup>st</sup>	–	–	–	Yuan <i>et al.</i> 2011
Ba	MG	AC	<i>Arundo d. active. C</i>	–	Ground	11, >5	(30)	L	9	2 <sup>nd</sup>	–	En	–	Zhang <i>et al.</i> 2008

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Ac	AGB	–	CMC gel	–	–	< 4	–	–	40-90	–	–	–	Uptake was correlated with CMC content of gel.	Abou-Taleb <i>et al.</i> 2009
Ac	AR57	F	<i>Neurospora crassa</i> cells	H	ground	1	Low	All	36-48	2 <sup>nd</sup>	–	Ex	–	Akar <i>et al.</i> 2006
Ac	AB40	W	<i>Thuja orient cones</i>	H	crush	1	Low	L	97	2 <sup>nd</sup>	–	Ex	–	Akar <i>et al.</i> 2008
Ac	AR274	PI	<i>Dicranella v moss</i>	H	blender	3	30	L	160-2000	2 <sup>nd</sup>	–	Ex	Not affected by stirring	Akkaya & Özer 2005
Ac	Metal compl.	F	<i>Rhizopus a. fungus</i>	H	Homog.	2	–	RP, LF	61-370	–	–	–	Salt decreased sorption.	Aksu & Balibek 2010
Ac	AK 194	F	3 fungi	H	Homog.	1-2	25-35	L	667	2 <sup>nd</sup>	–	Var	Internal diffusion; Kinetic fit aslo OK with first order model	Aksu & Karabayir 2008
Ac	Metal compl.	B	<i>Phormidium</i>	H	–	2	Low	L	91	2 <sup>nd</sup>	–	Ex	With Cr(VI)	Aksu <i>et al.</i> 2009
Ac	Metal compl.	F	Various fungi	H	Homog.	1-2	25-35	L	50-667	2 <sup>nd</sup>	–	En, Ex	–	Aksu & Karabayir 2008
Ac	AR151, Or2	F	<i>Aspergillus niger</i>	N	Homog.	(neut)	(30)	–	–	–	–	–	Shaking enhanced uptake.	Ali <i>et al.</i> 2008
Ac	AV	F	<i>Penicillium sp.</i>	N	–	5	35	L, F	6	1 <sup>st</sup>	–	–	No biodegradation, only bioadsorption	Anjaneya <i>et al.</i> 2009

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Ac	MO	Ag	Banana & orange peel	H	Crush	6-8; >6	–	F, L	16-21	–	–	–	Diffusion limits rates	Annandurai <i>et al.</i> 2002
Ac	CR	Ag	Banana & orange peel	H	Crush	6-8; >6	–	F, L	5-9	–	–	–	Diffusion limits rates	Annandurai <i>et al.</i> 2002
Ac	AmK	Ag	Banana & orange peel	H	Crush	6-8; >6	–	F, L	2-4	–	–	–	Diffusion limits rates	Annandurai <i>et al.</i> 2002
Ac	AB25	AC	Waste tea AC	–	–	7	(30)	–	203	–	–	–	–	Auto & Ha-meed 2011
Ac	CR	Tx	Jute proc. waste	H	–	Low	–	L, F	13	–	–	–	–	Banerjee & Dastidar 2005
Ac	AR	PI	<i>Pyracantha coccinea</i>		Cationic surfact.	2	Low	L	105	2 <sup>nd</sup>	–	Ex.	Diffusion control; salt slightly inhibits sorption	Akar <i>et al.</i> 2010
Ac	AB161	F	<i>Trametes versicolor</i>	H	with ch-itosan	3	45	L	180-207	2 <sup>nd</sup>	–	En.	Pore diffusion	Aksu <i>et al.</i> 2008
Ac	AB90	AC	<i>Paulownia</i> wood	–	–	–	–	F,L	–	–	O-cont. groups	–	–	Alam <i>et al.</i> 2009
Ac	AR151	F	<i>Aspergillus n.</i>	N	Homog.	–	(30)	–	–	–	–	–	Fungi tolerant of 10-1000 ppm of dye.	Ali <i>et al.</i> 2008
Ac	AR183 AG25	N	<i>Pistacia k</i> shells	H	Crush	2	(35)	L	7-33	2 <sup>nd</sup>	–	En	–	Aydin & Baysal 2006
Ac	AY29	AC	<i>Paulownia tomentosa</i>	–	KOH, acids	–	–	–	–	1 <sup>st</sup>	Less O	En	–	Bangash & Alam 2006

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Ac	AB1	AC	<i>Ailanthus altissima</i>	–	–	–	–	–	–	1 <sup>st</sup>	Lo carbonyl	–	–	Bangash & Alam 2006
Ac	4 dyes	Tx	Cationized cotton	–	–	–	Low	L	250-800	–	–	Ex	–	Baouab <i>et al.</i> 2000
Ac	3 dyes	W	Cationized sawdust	D	Extrac, deriv.	–	Low	L	103-412	–	–	Ex	–	Baouag <i>et al.</i> 2001
Ac	AB25	Ag	<i>Litchi chinensis</i>	–	–	–	–	L	200	2 <sup>nd</sup>	–	–	–	Bhatnagar & Minocha 2010
Ac	CR	F	<i>Trametes versicolor</i>	A	–	Low	–	L, T	40-53	2 <sup>nd</sup>	–	–	Favored by autoclaving	Binupriya <i>et al.</i> 2007
Ac	15 dyes	Tx	Cotton	–	–	No effect	–	–	–	–	–	Var .	Binding enthalpy due to van der Waals	Bird <i>et al.</i> 2006
Ac	RY23	Tx	Cotton	–	Grafted quats, levels	–	–	–	100-300	–	–	–	Moderate grafting with quats increased uptake	Bouzaida & Rammah 2002
Ac	MO	AC	Hickory husk	–	ZnCl <sub>2</sub>	–	–	–	900	–	–	–	Optimized activation	Cao <i>et al.</i> 2010
Ac	CR		Hazelnut shells	H	Support for biota	–	(30)	–	–	–	–	–	Biodegradation by <i>Phanercha</i> .	Carletto <i>et al.</i> 2008
Ac	MB	AC	Ash from rice husk	–	As is	Low	(30)	RP	7	–	–	–	–	Chowdhury <i>et al.</i> 2009

Ac	AY34	Sl	Sewage sludge	–	With aeration	–	–	–	(%s)	–	–	–	Test procedure	Churchley <i>et al.</i> 2000
Ac	AB225, AB062	B	<i>Paenibacil. macerans</i>	H	–	1	–	L, T	95	2 <sup>nd</sup>	–	–	IEP pH=4	Colak <i>et al.</i> 2009
Ac	4 dyes	Ag	<i>Stipa tenacissima</i> L.	–	Cationic grafting	(7)	–	L	250-830	–	–	–	–	El Ghali <i>et al.</i> 2010
Ac	7 dyes	Ag	Guava seed	H	Not crushed	(6)	(25)	–	0.2-1.4	–	–	–	Note: an unfavorable pH value was used.	Elizalde-G. & H. 2009a
Ac	AB25	N, W	Hazelnut sh Sawdusts	H	ground	–	(20)	L	55	2 <sup>nd</sup>	–	–	–	Ferrero 2007
Ac	AB29	F	<i>Aspergillus niger</i>	A	H <sub>2</sub> SO <sub>4</sub> ground	4	–	L, etc.	6-14	1 <sup>st</sup> or 2 <sup>nd</sup>	–	–	Dead better than viable	Fu & Viraraghavan 2001b
Ac	AB29	F	<i>Aspergillus niger</i>	A	H <sub>2</sub> SO <sub>4</sub> ground	(4)	–	–	17-18	–	–	–	Functional groups control adsorption	Fu & Viraraghavan 2002a
Ac	CR	F	<i>Aspergillus niger</i>	A	HCO <sub>3</sub> ground	(6)	–	–	7-16	–	–	–	Functional groups control adsorption	Fu & Viraraghavan 2002a
Ac	CR	F	<i>Aspergillus niger</i> , and AC	A	HCO <sub>3</sub> ground	6	–	RP	12-15	2 <sup>nd</sup>	–	–	Autoclaving (killing) helped sorption	Fu & Viraraghavan 2002b
Ac	AY17	Sl	Aerobic gran sludge	H	–	2	(20)	T F, L	30-50	2 <sup>nd</sup>	N, OH, COOH	Ex	Diffusion control	Gao <i>et al.</i> 2010
Ac	AY17, AO7	AC	<i>Euphorbia macrocla.</i>	–	K <sub>2</sub> CO <sub>3</sub>	2	55	L	161, 455	2 <sup>nd</sup>	–	En	Desorp at pH 11.	Gercel & G 2009
Ac	AO7	Ash	Bottom ash, power plant	–	–	2	50	L	4	1 <sup>st</sup>	–	En	–	Gupta <i>et al.</i> 2006
Ac	AB49, AO13	AC	–	–	–	Any	25	L	11-14, 4-5	2 <sup>nd</sup>	–	Ex	–	Gupta <i>et al.</i> 2007
Ac	Carmoisine A	Ash	Bottom ash, power plant	–	–	2	(50)	L	28	2 <sup>nd</sup>	–	En	Regen. with NaOH	Gupta <i>et al.</i> 2009



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Ac	Indigo-carmin	Ag	Mustard, charcoal	H, —	Ground	Any, 2	30	L	—	—	—	Ex	Glycerol to desorb dye.	Gupta <i>et al.</i> 2010
Ac	AB113	AC	Rubber tire; Commercial	—	—	2, 5	25	L	9, 7	1st	—	Ex	—	Gupta <i>et al.</i> 2011
Ac	AG25	Ag	Durian peel	—	—	—	(30)	—	63	—	—	—	—	Hameed & Hakimi 2008
Ac	AG25	AC	Activ. Ash from palm	—	H <sub>2</sub> SO <sub>4</sub>	2	50	F	123-182	2 <sup>nd</sup>	—	En	The pH dependency was surprisingly weak.	Hameed <i>et al.</i> 2007a
Ac	PR, BrphB	AC	Act. charcoal	—	Fluka	low	low	L	18-24	—	—	Ex	pH effects depend on dye type	Iqbal & Ashiq 2007
Ac	Egacid O	W	<i>Picea</i> wood shavings	H	HCl, CO <sub>3</sub> Na <sub>2</sub> HPO <sub>4</sub>	high	—	L-F	19-220	—	—	—	Acid treatment helped EO sorption.	Janoš <i>et al.</i> 2009
Ac	AY49	AC	Pyrolysis char sludge	—	N <sub>2</sub> or CO <sub>2</sub>	(6-7)	—	L	116	—	—	—	High-temp treatment increased sorption	Jindarom <i>et al.</i> 2007
Ac	AB25	AC	Act. C from plum kernel	—	Steam	—	(30)	—	70-250	2 <sup>nd</sup>	—	—	Higher temp of prep helped; diffusion control	Juang <i>et al.</i> 2000

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Ac	AB120	F	<i>Aspergillus lentulus</i>	L	Nutritional condition	(6.5)	(30)	–	–	–	–	–	Glucose greatly increased removal.	Kaushik & Malik 2010
Ac	AB120	F	<i>Aspergillus lentulus</i>	L	Glucose & urea	(6.5)	(30)	–	–	–	–	–	Nutrient optimization: glucose + urea	Kaushik & Malik 2011
Ac	AY99	PI	Coir pith	H	crush	2	(30)	L-F	442	–	FTIR shifts	–	Complexation & electrostatic	Khan <i>et al.</i> 2011
Ac	IC	–	–	–	–	5	(30)	–	34	–	–	–	–	Lakshmi 2009
Ac	AB2	Ma	<i>Eichhornia crassipes</i>	–	–	Low	–	L	108	–	–	–	Diffusion	Lee <i>et al.</i> 1999
Ac	DR	Ag	<i>Schizophyl.</i> + rice hull	L, A	Fungus on hull	–	(30)	–	–	–	–	–	Both sorption & enzymatic	Li & Jia 2008
Ac	AR Supran.	Al	<i>Chlorella vulgaris</i>	L	–	–	–	L, F	256	–	–	–	Biosorption little affected by nutrients	Lim <i>et al.</i> 2010
Ac	AR183	AC	Act. C	–	KOH act, CO <sub>2</sub>	–	(26)	–	400-2700	2 <sup>nd</sup>	–	–	Diffusion control	Ling <i>et al.</i> 2011
Ac	AO8 AB45	F	<i>Penicillium chrysogen.</i>	D	PEI x-l. glutarald	low	(30)	F, L	33, 18; 352, 196	–	FTIR, amine	–	Little effect of temperature.	Low <i>et al.</i> 2008
Ac	AR18 AR27	Ce	Cellophane membrane	N	–	–	(25)	F	36	–	–	–	Diffusion rates; salt promoted sorption.	Maekawa & Kondo 1996

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Ac	Rhod, Sandol	Ag	Bark, rice husk, cotton	H	–	–	(30)	–	0, 15-20	–	–	–	Bark > cotton > rice husk	McKay <i>et al.</i> 1986
Ac	AY36	AC	Act. C from sawdust, rice husk	–	Steam act.	3	(30)	L, F	184, 87	1 <sup>st</sup>	–	–	Diffusion control.	Malik 2003
Ac	AO10	FA	Fly ash fr. bagasse	–	Sieving	4	(30)	R-P	19	2 <sup>nd</sup>	–	–	–	Mall <i>et al.</i> 2006a
Ac	AB25, AY117	AC	Bamboo char	–	Crush	–	–	R-P	17, 29	–	–	–	–	Mui <i>et al.</i> 2010
Ac	CR, Procion o	Ag	Orange peel	D	Crush	Low	(29)	L, F	22, 1	1 <sup>st</sup>	–	–	Desorption at alkaline pH	Namasivaya m <i>et al.</i> 1996
Ac	Brilliant B	Ag	Banana pith	D	Ground	Low	(30)	L, F	4	1 <sup>st</sup>	–	–	–	Namasivaya m <i>et al.</i> 1998
Ac	CR	PI	<i>Diospyros melan.</i> leaf	–	H <sub>2</sub> O <sub>2</sub>	(6)	–	F, L	47-134	–	–	–	Peroxide incr. sorption.	Nagda & Ghole 2009
Ac	Amido black	AC	Act. C from palm flower	–	H <sub>2</sub> SO <sub>4</sub>	2	(27)	F	2-4	2 <sup>nd</sup>	–	En	Sample prep not well described.	Nethaji & S. 2011
Ac	AB40	Ag	Barley straw	H	Cetylpyr chloride	–	(25)	L	48	–	–	–	Cat surfactant helped	Oei <i>et al.</i> 2009
Ac	“BDH”	Ag	Palm kernel fiber	H	Ground	(5)	25	L	39	–	–	Ex	–	Ofomaja & Ho 2007
Ac	CR	Ag	Palm kernel seed coat	H	NaOH, Ground	(6.7)	–	L	66	2 <sup>nd</sup>	–	–	Interparticle diffusion	Oladoja & A 2009
Ac	AR274	Ma	<i>Enteromorpha prolifera</i>	H	Blender	2-3	30	L	244	2 <sup>nd</sup>	–	Ex	–	Ozer <i>et al.</i> 2005
Ac	AB290 AB324	Al	<i>Spirogyra rhizopus</i>	H	Blender	2-3	25-30	L	1360, 367	2 <sup>nd</sup>	–	Ex	–	Ozer <i>et al.</i> 2006

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Ac	AR88, AG3, AO7	Ma	<i>Azolla filic-uloides</i> alga	D	Crushed	7, 3, 3	–	L, F	109, 133, 110	2 <sup>nd</sup>	–	–	–	Padmesh <i>et al.</i> 2006a
Ac	AB15	Ma	<i>Azolla filic-uloides</i> alga	D	Crushed	7,	–	L	36-116	2 <sup>nd</sup>	–	–	Column method sorbed much less.	Padmesh <i>et al.</i> 2006b
Ac	AR88	Ma	<i>Azolla microphylla</i>	–	–	3	30	L, Khan	55	–	–	–	–	Padmesh <i>et al.</i> 2006c
Ac	AO7, 3 others	AC	Activated C	–	HNO <sub>3</sub> , H <sub>2</sub> , O <sub>2</sub>	5-7	–	–	–	–	–	–	Attributed to redox breakdown	Pereira <i>et al.</i> 2010
Ac	AB25	W	Spruce wood	–	Ground	–	–	–	–	–	–	–	Used to estimate surface area	Poots <i>et al.</i> 1976
Ac	AR87	AC	Activated C from jute fib	–	H <sub>3</sub> PO <sub>4</sub>	2	30	L, F	18	2 <sup>nd</sup>	–	–	Surface & pore diffusion	Porkodi & Kumar 2007
Ac	AB9	Ma	<i>Tubinaria conoides</i>	H	HCl	1	33	F, RP	22	2 <sup>nd</sup>	–	–	–	Rajeshkanna <i>n et al.</i> 2010
Ac	AB5	–	<i>Eichhornia crassipes</i>	–	–	–	–	–	–	–	–	–	Packed bed optimization	Ramasamy <i>et al.</i> 2008
Ac	AB62 AR266 AY49	F	<i>Cunninghamella ele-ans</i>	A	Lyophilized	–	(25)	–	300-600	1 <sup>st</sup> & 2 <sup>nd</sup>	–	–	Competition among the 3 dyes.	Russo <i>et al.</i> 2010
Ac	AB9	F	<i>Trichoderm. harzianum</i>	N	–	2-4	(30)	L, F	3	2 <sup>nd</sup>	–	–	–	Sadhasivam <i>et al.</i> 2007b

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Ac	AV 17	Ce	Cotton fiber	H	Methanol extracted	8.5	(24)	—	13-25+	—	—	En	Heterogeneous	Saleem <i>et al.</i> 2007
Ac	AB 25	—	Cellulose, amidoxyl	—	Amidoxylated	—	(25)	—	550	—	Ternary	—	Metals involved in dye sorption	Saliba <i>et al.</i> 2002
Ac	CR	N	Cashew nut shells	—	—	(3)	(30)	—	5	—	—	—	—	Senthil K. <i>et al.</i> 2010
Ac	MO	F	16 fungi	N, A	—	—	(30)	—	—	—	—	—	Live spores more effective	Seyis & Suba-sioglu 2008
Ac	AR18	AC	<i>Populus</i>	—	H <sub>3</sub> PO <sub>4</sub>	Low	—	L	4	1 <sup>st</sup>	—	—	—	Shohoochi <i>et al.</i> 2010
Ac	AO	F	<i>Aspergillus niger</i>	—	—	2	—	L, F	0.5	2 <sup>nd</sup>	—	—	—	Sivasamy & S 2011
Ac	AB 25 AR 151	AC	Activated C from wood	—	H <sub>3</sub> PO <sub>4</sub>	2-3	(21)	L	555-588	—	—	—	Loss of functional groups on heating	Tsang <i>et al.</i> 2007
Ac	AR57	—	<i>Phaseolus vulgaris</i> L.	—	—	—	(20)	L	215	2 <sup>nd</sup>	—	Ex	—	Tunali <i>et al.</i> 2007
Ac	AB80	AC	Bagasse	—	ZnCl <sub>2</sub> , Mg, Ca	—	—	—	50-500	—	—	—	Pore models	Valix <i>et al.</i> 2006
Ac	AV17	AC	Pistachio nut shell	—	H <sub>2</sub> SO <sub>4</sub>	2	>45	L	79-125	2 <sup>nd</sup>	—	En	—	Vijayalakshmi <i>et al.</i> 2011

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Ac	Tectilon B	B	3 bacteria	N	—	(7)	(25)	—	—	—	—	—	19% sorbed in aerobic biodegrad.	Walker & W. 2000
Ac	CR	Ag	Wheat bran, Rice bran	H	Ground	(8)	(25)	L, F	23, 15	2 <sup>nd</sup>	—	—	—	Wang & Chen 2009a
Ac	CR	Al	<i>Porphyra yezoensis</i>	H	—	—	(25)	L, F	71	2 <sup>nd</sup>	—	—	—	Wang & Chen 2009b
Ac	AB74	AC	Fir wood char	—	KOH, CO <sub>2</sub>	—	(30)	L	190-350	—	—	—	—	Wu & Tseng 2006
Ac	AO7	C	Hydroxypropyl cellulose	—	Epichlorohydrin	4	—	L, F	2480	2 <sup>nd</sup>	—	—	—	Yan <i>et al.</i> 2009a
Ac	AB92	Ag	Wheat straw	—	Cationic, quat.	—	—	L, F	1072	2 <sup>nd</sup>	—	—	—	Yan <i>et al.</i> 2009b
Ac	AB9	AC	<i>Cannibus sativa</i> bast	—	H <sub>3</sub> PO <sub>4</sub>	—	(20)	L	29	—	—	—	—	Yang 2011a
Ac	AK172	F	<i>Penicillium YW</i>	A, H	Ground	3	40	L	225	2 <sup>nd</sup>	Groups present	—	Intraparticle diffusion	Yang 2011b
Ac	AB74	AC	Fir wood char	—	NaOH	—	(30)	L	700-900	—	—	—	Surface coverage	Wu & Tseng 2008
Ac	AB255, AB62	B	<i>Bacillus amyloliquef</i>	—	—	1	—	—	111, 112	2 <sup>nd</sup>	—	—	—	Yenikaya <i>et al.</i> 2010
Ac	BPhB	F	4 fungi	A, H	NaOH, ground	2	(25)	L	526-1111	2 <sup>nd</sup>	—	—	—	Zeroual <i>et al.</i> 2006a
Ac	BPhB	F	<i>Rhizopus stolonifer</i>	A, H	NaOH, or HCO <sub>3</sub> , ground	(2)	(25)	L	333-1111	2 <sup>nd</sup>	—	nul	Regenerated with NaOH	Zeroual <i>et al.</i> 2006b

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Dir	DB71	FA	Palm ash	N	–	–	(30)	L, F	400	2 <sup>nd</sup>	–	–	–	Ahmad <i>et al.</i> 2007
Dir	DB1	Ce	Cellophane; Cotton	–	–	(7.2)	–	–	–	–	–	–	H-bonds with cellulose; apolar environment	Abbot <i>et al.</i> 2004a
Dir	Pink	–	CMC gel	–	–	< 4	–	–	40-140	–	–	–	Uptake was correlated with CMC content of gel.	Abou-Taleb <i>et al.</i> 2009
Dir	–	–	–	–	–	–	–	–	–	–	–	–	–	Ahmad <i>et al.</i> 2007
Dir	DB106	AC	Pomegranat peel AC	–	–	2	(25)	–	43-58	–	–	–	–	Amin 2009
Dir	2 DRs	Ag	Orange peel	D	Crush	2	25	L	11-21	2 <sup>nd</sup>	–	–	Optimum pH could have been yet lower.	Arami <i>et al.</i> 2005
Dir	DR 80	N	Almold shells	–	–	Weak effect	20	L	16-21	2 <sup>nd</sup>	–	–	–	Ardejani <i>et al.</i> 2008
Dir	DY106, DRY, DB25	–	Bleached cotton	–	–	–	–	L	25, 20, 20	–	–	–	No salt used; competition shown	Bairathi 1993
Dir	2 DRs	F	<i>Trametes versicolor</i>	N, H	–	6, 3	–	F, T	101-225	2 <sup>nd</sup>	–	–	Benzidine-based dyes on WRF; heat treatment helped	Bayramoglu & Arica 2007

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Dir	DB1	Ce	Lyocell	D	–	–	(80-120)	F	8	–	–	–	Fabric dying, 0.17 M salt	Carillo <i>et al.</i> 2002
Dir	DR75	Ma	<i>Posidonia</i>	H	Grafted, succinic	2	–	L, J	82-147	2 <sup>nd</sup>	–	–	Lead-saturated bio-mass effective for dye	Chadlia <i>et al.</i> 2009
Dir	DR80	Sl	Sewage sludge	–	With aeration	–	–	–	(%s)	–	–	–	Test procedure	Churchley <i>et al.</i> 2000
Dir	DO8 DB2	W	Oak sawdust	–	As is	3, 4.5	(25)	L	6,9	1 <sup>st</sup>	–	–	–	Dullman <i>et al.</i> 2002
Dir	DN dyes	W	Beech sawust	–	Milled	Low	–	L	526	2 <sup>nd</sup>	–	–	–	Dulman & C 2009
Dir	DB86	AC	Act. C from orange peel	–	H <sub>2</sub> SO <sub>4</sub>	2	(25)	L, T	34	2 <sup>nd</sup>	–	–	–	El Nemr <i>et al.</i> 2009
Dir	DB2	W	Wood pulp	–	DMA-epi rx	–	–	L, F	102	–	–	–	–	Hashem 2006
Dir	DB86, DR83	Ce	Alpha cellulose	–	Rx adipic, diethtri-amine Epichlo.	–	–	–	–	–	–	–	Improved sorptive capacity	Hwang & Chen 1993
Dir	DB71	Ce	Lyocell cellulose	H	NaOH	–	(80)	–	15 to >>20	–	–	–	Salt helped, fitting Donnan theory	Ibbett <i>et al.</i> 2007a



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Dir	DB71	Ce	Lyocell cellulose	H	UF resin	–	(25)	–	15-30	–	–	–	Resin hurt dye sorp, but NaOH treatment overcame this effect.	Ibbett <i>et al.</i> 2007b
Dir	DR, Ac, Rx dye	W	Wood sawdust	D	PEI	4.5-8	(25)	–	–	–	–	–	Amination greatly increased sorption	Ibrahim <i>et al.</i> 1997
Dr.	Various pairs	Ce	Bleached cotton yarn	D	Proton, oxidized	(6.5)	(75)	–	2-20	–	–	–	Competition shown	Johnson <i>et al.</i> 1974
Dir	DB71	Ce	Regen. celluloses	–	–	–	50 - 100	–	2-6	–	–	–	Dyes to characterize accessibility	Kreze <i>et al.</i> 2002
Dir	DY28	Tx	Cotton	–	NaOH, deterg., H <sub>2</sub> O <sub>2</sub> , cat .	(4-7)	(40-95)	–	–	–	–	–	Dye increased neg. zeta potential of cotton.	Kumar & Teli 2007
Dir	DB 2B DG B	W	Sawdust	–	–	(3)	(30)	–	496, 298	–	–	–	–	Malik 2004
Dir	CR, Solr B	Ag	Bark, rice husk, cotton	H	–	–	(30)	–	0, 0	–	–	–	Bark > cotton > rice husk	McKay <i>et al.</i> 1986
Dir	DN1	Al	<i>Spirogyra</i> sp. I02	D	–	Low	30	L	5.5	2 <sup>nd</sup>	–	–	Intra-particle diffusion fit well	Mohan <i>et al.</i> 2008

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Dir	Brilliant B	Ag	Banana pith	D	Ground	Low	(30)	L, F	6	1 <sup>st</sup>	–	–	–	Namasivayam <i>et al.</i> 1998
Dir	–	Ma	<i>Posidonia oceanica</i> L	–	–	–	(30)	F	3	–	–	En	–	Ncibi <i>et al.</i> 2006
Dir	DO8, DB6, DO37, DN2, DG26, DN95, DOR	W	Beech wood flour	–	–	(3) Optimum from prev work	(22)	–	–	–	-NH, -OH	–	FTIR, DTA show H bonding involving the –NH and –OH groups of the dyes with the O sites on the cellulose.	Odochian <i>et al.</i> 2007
Dir	DB15, 8 other	F	<i>Phanerochaete chrysosporium</i>		Mn peroxidase	(5)	(39)	–	–	–	–	–	Enzymatic decay of DB15 only; sorption also important	Pazarlioglu <i>et al.</i> 2005
Dir	FWA	F	<i>Aspergillus awamori</i>	N	–	2.5	(30)	–	–	–	–	–	Biosorption mostly & biodegradation	Rao & Venkateswarlu 2006
Dir	DR31 DO26	Ag	Rice husk	–	HCl, alginate	–	–	–	46-67	2 <sup>nd</sup>	several	En	–	Safa <i>et al.</i> 2011
Dir	DR 80	Tx	Cotton fiber	H	Methanol extracted	8.5	(24)	–	60-80	–	–	En	Heterogeneous	Saleem <i>et al.</i> 2007

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Dir	FWA	Tx	Bleached cotton	–	–	–	–	–	–	–	–	–	Fluorescence shows mechanism.	Stana <i>et al.</i> 1995
Dir	DB199	F	<i>Aspergillus niger</i>	H	Ground	Low	45	L, F	30	2 <sup>nd</sup>	–	En	–	Xiong <i>et al.</i> 2010
Rx	V	–	CMC gel	–	–	< 4	Low	–	20-80	–	–	–	Uptake was correlated with CMC content of gel.	Abou-Taleb <i>et al.</i> 2009
Rx	RR45	F	<i>Agaricus b</i> & <i>Thuja o</i> fungal mix	H	Crush	1-2	Hi	–	45-109	2 <sup>nd</sup>	Yes	–	Endothermic; no effect of metal ions; functional group interactions	Akar <i>et al.</i> 2008a
Rx	RR2	F	Fungal	H	Crush, CTAB	Low	Hi	L	142	2 <sup>nd</sup>	Yes	En	Removal increased by CTAB	Akar & Divriklioglu 2010
Rx	RR198	F	<i>Aspergillus parasiticus</i>	–	–	2	Hi	L,F, DR	40	–	–	En	–	Akar <i>et al.</i> 2009a
Rx	RB49	F	Fungus & <i>Thuja</i> cone	H	Crush	1	Hi	–	3-8	2 <sup>nd</sup>	–	En ?	IEP pH=2	Akar <i>et al.</i> 2009b
Rx	RR198	Ag	Olive pomace	H	Crush	2	Hi	L	39-43	2 <sup>nd</sup>	COOH	En	Weak competition by salt	Akar <i>et al.</i> 2009f

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Rx	RR195	W	<i>Pinus sylv. cones</i>	H	Crush	1-2	Hi	F, L	4-7	2 <sup>nd</sup>	–	En	–	Aksakal & Uzun 2010
Rx	3 dyes	F	<i>Rhizopus a fungus</i>	H	Ground	2	–	–	284-1008	–	–	–	Breakthrough curves; results affected by flow	Aksu <i>et al.</i> 2007a
Rx	3 dyes	F	<i>Rhizopus a fungus</i>	H	Ground	(2)	(25)		636-1008	–	–	–	Flow & concentration favored uptake; breakthrough curves	Aksu <i>et al.</i> 207b
Rx	Rema-Zol Blue	F	Yeasts	H	–	2	(25)	L	250	2 <sup>nd</sup>	–	–	–	Aksu & Donmez 2002
Rx	Gema-zol tbG	Ag	Sugar beet pulp	H	Ground	2	25	L	235	Var	–	Ex	Surface & pore diffusion	Aksu & Isoglu 2006
Rx	RK5	F	<i>Rhizopus a fungus</i>	H	Blender	2	35	F	50-300	2 <sup>nd</sup>	–	–	–	Aksu & Tezer 2000
Rx	3 dyes	Al	<i>Chlorella vulgaris</i>	H	–	2	25-35	Var	420	2 <sup>nd</sup>	–	Ex.	–	Aksu & Tezer 2005
Rx	3 dyes	AC	Act. C	–	–	(4-5)	(20)	L, R-P	65-714	–	–	–	Competitive adsorption	Al-Degs <i>et al.</i> 2007
Rx	3 dyes	AC	Act. C	–	Comm-ercial	(7)	(25)	L,F	210	–	–	En.	Considers numbers of functional groups	Al-Degs <i>et al.</i> 2008

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Rx	Drimar-ine blu	F	<i>Aspergillus niger</i>	N	Homog.	(neut)	(30)	–	–	–	–	–	Shaking enhanced uptake.	Ali <i>et al.</i> 2008
Rx	RO	Ag	Pith of sugar bagasse	–	–	1	(25)	–	2-4	–	–	–	–	Amin 2008
Rx	Verofix red	–	Chitin	–	Comm-ercial	weak func.	20	–	38	–	–	–	Response surface	Annadurai & Sheeja 1998
Rx	RR120	F	<i>Leninus sajor-c</i> fung	N, H	Acid, alkali	3, Low	–	L	57-183	2 <sup>nd</sup>	–	–	Heat treat increased uptake; acid or alkali treatments did not.	Arica & Bayramoglu 2005
Rx	RY42, RR45	Ag	<i>Citrus sinensis</i>	D	Acetic, a-nitrile	2	–	F	8-17	2 <sup>nd</sup>	Tried	–	Substantial desorption, presumably into water.	Asgher & Bhatti 2010
Rx	RB19	SI	Pulp & pap sludge	–	–	3	–	T	33	2 <sup>nd</sup>	–	–	–	Azizi <i>et al.</i> 2010
Rx	RB19	–	Wood waste	–	–	2	–	L	31	2 <sup>nd</sup>	–	–	–	Azizi <i>et al.</i> 2011
Rx	RR120, Red E	Ce	Cellulose film	–	–	(6.8)	(80)	–	–	–	–	–	Salt helped adsorption; chemical potential	Bae <i>et al.</i> 1997a
Rx	RR120, Red E	Ce	Cellulose film	–	–	(6.8)	(80)	–	–	–	–	–	Salt affected diffusion of dye	Bae <i>et al.</i> 1997b

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Rx	RR120, Red E	Ce	Cellulose film	–	–	High	(80)	–	–	–	–	–	pH effects consistent with dissociation	Bae <i>et al.</i> 1998
Rx	RR2	Ce	Cellulose	–	–	–	–	–	–	–	–	–	Molec. dynamics sym.; minimal interaction	Baird <i>et al.</i> 2008
Rx	Mix	F	<i>Phaneroch. chrysosp.</i>	N	Rinsed	2	40	L	10	–	–	–	Desorb at high pH.	Bakshi <i>et al.</i> 2006
Rx	RY23	W	Cationized sawdust	D	Extrac, deriv.	–	Low	L	142-249	–	–	Ex	–	Baouag <i>et al.</i> 2001
Rx	RB4	F	<i>Phaneroch. chrysosp.</i>	N, H	Acid, or base	3	–	F, T	212	–	–	–	Heat & acid treatments increased sorption.	Bayramoglu <i>et al.</i> 2006
Rx	Drimar-ene Red	F	<i>Aspergillus foetidus</i>	–	–	2.5	60	–	344	–	–	–	Regenerate with NaOH	Bidisha <i>et al.</i> 2006
Rx	RR2	AC	<i>Bokbunja</i> seed	–	Hexane extracted	2	–	–	30	1 <sup>st</sup>	–	–	–	Binupriya <i>et al.</i> 2008a
Rx	RB4	Ba	<i>Bacillus subtilis</i>	A	Exterior polysac	Low	–	L	16-43	1 <sup>st</sup>	FTIR groups	–	Immobilization hurt sorption.	Binupriya <i>et al.</i> 2010
Rx	Burazol B ED	SI	Anaerobic sludge	H	–	0.5	50	–	118-127	2 <sup>nd</sup>	–	SI. En	–	Caner <i>et al.</i> 2009
Rx	RR194	N	<i>Cupuassu</i> shell	–	–	2	(25)	–	38	–	–	–	–	Carsodo <i>et al.</i> 2011

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Rx	RR120	Ba	<i>Spirulina platensis</i>	–	–	–	–	–	8-18	–	–	–	Tolerance of the dye	Celekli <i>et al.</i> 2009
Rx	RB4	SI	Sewage sludge		With aeration	–	–	–	(%s)	–	–	–	Test procedure	Churchley <i>et al.</i> 2000
Rx	Procion R, V	F	<i>Aspergillus oryzae</i>	N, A	Pellets	2.5	–	–	–	–	–	–	Autoclaving helped	Corso & Almedia 2009
Rx	Remaz Rifacio.	FA	Fly ash	–	–	6	50	L, F	7-135	–	–	En	Diffusion control of removal	Dizge <i>et al.</i> 2008
Rx	RK5	AC	Act. C; fly ash	–	–	as-is	(20)	L, F	59, 8	2 <sup>nd</sup>	–	–	–	Eren & Acar 2006
Rx	RK5	FA	Fly ash	–	–	as-is	(20)	F	7	2 <sup>nd</sup>	–	–	–	Eren & Acar 2007
Rx	Remaz BrBBR	AI	<i>Scenedes. quadricaud.</i>	–	–	2	30	L,F, DR	44-95	2 <sup>nd</sup>	–	–	–	Ergene <i>et al.</i> 2009
Rx	Remaz RKB	B	Thermophil. cyanobact.	–	Immobil.	7-9	45	–	41	–	–	–	Live immobilized bacteria	Ertugrul <i>et al.</i> 2008
Rx	RR228	Ag	Flax shive	D	Ground, chitosan	2	–	L	14	2 <sup>nd</sup>	–	–	–	Feng <i>et al.</i> 2012
Rx	RB5	SI	Activated sludge	H		2	20	L	71-117	2 <sup>nd</sup>	–	Ex	Diffusion control	Gulnaz <i>et al.</i> 2006
Rx	Several	F	<i>Chara vulgaris</i>	–	–	–	–	–	11-35	2 <sup>nd</sup>	–	–	Intraparticle diffusion	Gulnaz 2009
Rx	RR198	–	<i>Potamogeton crispus</i>	–	Native, alkali	–	(20)	–	14-44	2 <sup>nd</sup>	several	–	lep = 3.8 to 4.6	Gulnaz <i>et al.</i> 2011

Dye Class	Dye	Biomass class	Biomass	Dried, never, heat	Modification	Best pH	Best temperature	Isotherm best fit	Adsorp. capac. (mg/g)	Rate law best fit	Functional groups	Thermodynamics	Key Findings	Author (year)
Rx	—	B	<i>Corynebacterium glut.</i>	—	—	—	—	—	—	—	Amino	—	COOH & phosphate inhibited sorption.	Han & Yun 2007
Rx	5 dyes	Ce	Lyocell cellulose	N, H	Drying or not	—	(25)	—	Partition ratios	—	—	—	Drying hurt sorption to different extents for different celluloses.	Ibbett <i>et al.</i> 2007a
Rx	RO122	F	<i>Penicillium restrictum</i>	H	ground	1	50	L	181-220	2 <sup>nd</sup>	—	En	—	Ilhan <i>et al.</i> 2008
Rx	RK5	AC	From bamboo	—	—	5	(25)	—	281, 442	—	—	—	—	Ip <i>et al.</i> 2011
Rx	Remaz BBR	F, Ag	<i>Phaneroch. chrysosp</i> ; Loofa alone	—	Free vs. immob.; -	2	(30)	L	85-101 6	2 <sup>nd</sup>	—	—	Loofa immob helped.	Iqbal & Saeed 2007
Rx	RK5	F	<i>Penicillium restrictum</i>	H	ground	1	50	—	98-112	2 <sup>nd</sup>	—	En	—	Iscen <i>et al.</i> 2007
Rx	RR198	SI	Pyrolysis char sludge	—	N <sub>2</sub> or CO <sub>2</sub>	(6-7)	—	L	25	—	—	—	High-temp treatment increased sorption	Jindarom <i>et al.</i> 2007
Rx	RhB	SI	Act. sludge	H	Acid, Alkaline, Control	—	—	L	7, 6 5	2 <sup>nd</sup>	—	—	Treatments all helped.	Ju <i>et al.</i> 2008
Rx	RO2/3	AC	C from cior pith	—	700 °C	Low	(35)	L	2.6	2 <sup>nd</sup>	—	—	Diffusion control	Kavitha & N 2007a



Dye Class	Dye	Biomass class	Biomass	Dried, never, heat	Modification	Best pH	Best temperature	Isotherm best fit	Adsorp. capac. (mg/g)	Rate law best fit	Functional groups	Thermodynamics	Key Findings	Author (year)
Rx	Synaz.	F	<i>Aspergilhis niger</i> fung.	A	Living or autocla.	3	30	–	–	–	–	–	Autoclaving helps sorption.	Khalaf 2008
Rx	5 dyes	Ma	<i>Lemna minor</i> duckweed	L	Triacon-tanol hormone	(6)	–	–	–	–	–	–	Bioremoval effective, tolerance to toxicity	Kilic <i>et al.</i> 2010
Rx	Burazol Blu ED		<i>Neurospor. crassa</i>	F	–	1	30	L	110	2 <sup>nd</sup>	–	En	–	Kiran <i>et al.</i> 2009
Rx	–	Tx	Cellulose fabric	–	KIO <sub>4</sub> , Chitosa n	–	–	–	–	–	–	–	Greatly enhanced dyeing	Kitkulnumcha i <i>et al.</i> 2008
Rx	5 dyes	F	5 fungal: <i>Aspergillus</i> , <i>Rhizopus</i>	A, H	powder	(6)	(29)	L, F	112-204	–	–	–	Anionic dyes	Kumari & Abraham 2007
Rx	RB2	Ma	<i>Eichhornia crassipes</i>	–	–	Low	–	L	59	–	–	–	Diffusion	Lee <i>et al.</i> 1999
Rx	RO16, RK5	AC	Act. C, 3 kinds	–	powder	–	(25)	–	150-550	–	–	–	Combination with alum & membrane.	Lee <i>et al.</i> 2006
Rx	RR141	W, FA, AC	Wood shv. bottom ash; Act. C	–	None; H <sub>2</sub> SO <sub>4</sub> ; –	10-11	(30)	F	24; 30; 42	–	–	En	–	Leechart <i>et al.</i> 2009
Rx	Levafix navy B	Al	<i>Chlorella vulgaris</i>	L	–	–	–	L, F	189	–	–	–	Biosorption little affected by nutrients	Lim <i>et al.</i> 2010
Rx	Lanaset R2GA	Al	<i>Chlorella vulgaris</i>	L	–	–	–	L, F	345	–	–	–	Biosorption little affected by nutrients	Lim <i>et al.</i> 2010

Dye Class	Dye	Biomass class	Biomass	Dried, never, heat	Modification	Best pH	Best temperature	Isotherm best fit	Adsorp. capac. (mg/g)	Rate law best fit	Functional groups	Thermodynamics	Key Findings	Author (year)
Rx	RR194, hydrol.	W	<i>Araucaria angustifol.</i>	H	Ground, Cr, acid & Cr	2	(25)	Sip, R-P	10	2 <sup>nd</sup>	–	–	–	Lima <i>et al.</i> 2008
Rx	Remaz BBR	F,B	Fungi & bacteria	L	–	–	(30-45)	–	–	–	–	–	Ligninase correlation	Lopez <i>et al.</i> 2006
Rx	Cibacron Red	Ma	–	–	H <sub>3</sub> PO <sub>4</sub> , HNO <sub>3</sub>	5-6	Hi	F	4-5	2 <sup>nd</sup>	–	En	–	Mahjoub <i>et al.</i> 2008
Rx	RR4	B	<i>Corynebact glutamicum</i>	D	As-is; PEI x-l.	(9)	(25)	L	172; 485	–	–	–	Desorption at pH 9	Mao <i>et al.</i> 2009b
Rx	RK5	F	<i>Funalia trogii</i>	L	Immobilized on <i>Luffa</i>	(6)	(30)	–	–	–	–	–	Laccase decolorization, not sorption	Mamanci and Ünyayar 2005
Rx	Two dyes	Ag	Esparto fiber with zeolite	–	NaOH, H <sub>2</sub> O <sub>2</sub> ; acrylics	>9	>70	–	120-320	–	–	En	–	Marzoug <i>et al.</i> 2011
Rx	Remaz.B B	W	<i>Eucalyptus</i> bark	D	–	2-2.5	Low	–	23	–	–	–	Salt helped sorption.	Morais <i>et al.</i> 1999
Rx	Alpacid gray	Ma	<i>Posidonia oceanica</i>	H	–	2	(30)	L	9	2 <sup>nd</sup>	–	–	–	Ncibi <i>et al.</i> 2008a
Rx	Alpacid gray	Ma	<i>Posidonia oceanica</i>	H	–	2	(30)	L, R-P	15	2 <sup>nd</sup>	–	–	–	Ncibi <i>et al.</i> 2008b
Rx	Remazol dyes	Ag	Straw, cob, wood	–	Chop	–	var	–	–	–	–	–	Dye helps CN ratio compost	Nigam <i>et al.</i> 2000

Dye Class	Dye	Biomass class	Biomass	Dried, never, heat	Modification	Best pH	Best temperature	Isotherm best fit	Adsorp. capac. (mg/g)	Rate law best fit	Functional groups	Thermodynamics	Key Findings	Author (year)
Rx	RR	W	Beech wood flour	–	–	(3) Optimum from prev work	(22)	–	–	–	-NH, -OH	–	FTIR, DTA show H bonds involving the –NH and –OH groups of the dyes with the O sites on the cellulose.	Odochian <i>et al.</i> 2007
Rx	RK5	Ag	Barley straw	H	Cetylpyr chloride	–	(25)	L	25	–	–	–	Cat surfactant helped	Oei <i>et al.</i> 2009
Rx	RO16 RR4, RB19	F	<i>Rhizopus arrhizus</i>	H	ground	2	–	–	200, 150, 100	–	–	–	No interference from cadmium; better than activated C	O'Mahony <i>et al.</i> 2002
Rx	RK5	F	<i>Aspergillus foetidus</i>	N	0.1 M NaOH	2-3	50	F	106	2 <sup>nd</sup>	–	En	Autoclaving helped; regenerate w NaOH.	Patel & Suresh 2008
Rx	RK5, RY176	FA	Fly ash fr. biomass	–	–	8.1-8.5	(25)	L	4, 3	2 <sup>nd</sup>	–	–	Little effect of salt	Pengthamk. <i>et al.</i> 2008
Rx	RK5	FA	Fly ash	–	NaOH	8-10	–	L	108	2 <sup>nd</sup>	–	–	–	Pengthamk. <i>et al.</i> 2010
Rx	RR	Ag	Rice husk	D	HNO <sub>3</sub>	Low	Hi	–	–	–	–	–	–	Ponnusami <i>et al.</i> 2007

Rx	10 dye mix	F	3 fungal, inactive	A	–	–	(30)	–	168-533	–	–	–	Column better than batch system.	Prigone <i>et al.</i> 2008
Rx	Carpet effluent	F	<i>Trametes versicolor</i>	L	Cultured	(4.5-5)	(21)	–	–	–	–	–	Effective with complex mix	Ramsay & Goode 2004
Rx	RK5 RB19	F	<i>Trametes versicolor</i>		Immob. alginate	(3.6)	(30)	–	–	1st	–	–	Not clearly sorption or breakdown.	Ramsay <i>et al.</i> 2005
Rx	Remazol Black B	F	<i>Aspergillus flavus</i>	L	Viable	4.5	30	–	4-233	–	–	–	Enhanced by glucose	Ramjusha <i>et al.</i> 2010
Rx	RR2	Ma	<i>Nymphaea rubra</i>	H	Crush	2	(30)	F	67	2 <sup>nd</sup>	Amine	–	Amine groups	Renganathan <i>et al.</i> 2009
Rx	5 dyes	Ag	Barley husks	H	–	(7.2)	(20)	L	8-20	–	–	–	–	Robinson <i>et al.</i> 2002a
Rx	5 dyes	Ag	Barley husks	H	–	(7.2)	(20)	L	2-9	–	–	–	Smaller particles more effective.	Robinson <i>et al.</i> 2002b
Rx	5 dyes	Ag	Apple pomace, wheat	H	–	–	(20)	L	2-8	–	–	–	Smaller particles more effective.	Robinson <i>et al.</i> 2002c
Rx	RK5	Ag	Potato peel	H	Ground	3	(25)	L, F	4	2 <sup>nd</sup>	–	–	–	Samarghandy <i>et al.</i> 2011
Rx	RO12, RR2, RB4	AC	Coir pith	–	H <sub>2</sub> SO <sub>4</sub>	1-3	(20)	F	–	1 <sup>st</sup>	–	–	–	Santhy & S. 2006
Rx	RR	AC	Activated carbon	–	H <sub>3</sub> PO <sub>4</sub>	Low	(28)	L	182, 200	–	–	–	Gas phase, poor resorption	Senthikumar <i>et al.</i> 2006a
Rx	RR120	Ce	Cellulose II, Lyocell	–	NaOH	Very high	(70)	L, F	42	–	–	–	Alkali swelling favors sorption	Siroky <i>et al.</i> 2011
Rx	3 dyes	F	<i>Aspergillus foetidus</i>	N	Acclimate	(8)	(30)	–	–	1 <sup>st</sup>	–	–	–	Sumathi & Manju 2000
Rx	Brill. R HE-3B	Ce	Cellulose derivatives	–	1° amine Quat.	<6 <10	55	–	350-6500	–	–	En	–	Suteu <i>et al.</i> 1999
Rx	Remazol blue	F	Fungus	–	–	5-6	–	–	–	–	–	–	–	Tastan <i>et al.</i> 2010

Dye Class	Dye	Biomass class	Biomass	Dried, never, heat	Modification	Best pH	Best temperature	Isotherm best fit	Adsorp. capac. (mg/g)	Rate law best fit	Functional groups	Thermodynamics	Key Findings	Author (year)
Rx	RB MR	AC	Cotton hull	–	Ground	2	(30)	L, F	13	2 <sup>nd</sup>	–	–	–	Thangamani <i>et al.</i> 2007
Rx	RR 23	AC	Activated C from wood	–	H <sub>3</sub> PO <sub>4</sub>	2	(21)	L	170	–	–	–	Loss of functional groups on heating	Tsang <i>et al.</i> 2007
Rx	RK5	B	<i>Corynebact glutamicum</i>	H	Decarbx, immob.	4	(25)	L	257	–	–	–	Intraparticle diffusion; regen. NaOH	Vijayaraghavan & Yun 2007a
Rx	RK5	B	<i>Corynebact glutamicum</i>	H	HNO <sub>3</sub> , other	1	35	L	195-419	2 <sup>nd</sup>	–	En	Regen. NaOH	Vijayaraghavan & Yun 2007b
Rx	RR4, RO16	B	<i>Corynebact glutamicum</i>	H	Immobil.: p-sulfone	2-3	–	L	43-180	–	Amine, etc.	–	Competition for sites	Vijayaraghavan & Yun 2008a
Rx	RK5	Al	<i>Laminaria</i> sp.	H	–	Low	–	F, L	101	–	Amine, etc.	En	Regen. NaOH	Vijayaraghavan & Yun 2008c
Rx	RK5	B	<i>Corynebact glutamicum</i>	H	Immobil.: p-sulfone	Low	–	–	69-87	–	Amine, etc.	En	Packed bed; regen NaOH	Vijayaraghavan & Yun 2008d
Rx	RK5	B	<i>Corynebact glutamicum</i>		Immobil.: alginate, p-sulfone	1	(25)	L	291-352	–	–	–	Intraparticle diffusion control; regen. with base	Vijayaraghavan <i>et al.</i> 2007
Rx	Mixture 4 dyes	B	<i>Corynebact glutamicum</i>		Decarbx, immob.	2-5	(25)	F	–	–	–	–	Decarboxylation very good; NaOH regen.	Vijayaraghavan <i>et al.</i> 2008a

Dye Class	Dye	Biomass class	Biomass	Dried, never, heat	Modification	Best pH	Best temperature	Isotherm best fit	Adsorp. capac. (mg/g)	Rate law best fit	Functional groups	Thermodynamics	Key Findings	Author (year)
Rx	RK5 & RO16	B	<i>Corynebact glutamicum</i>	H	Esterific; immob.: p-sulfone	1-4	(25)	L	248, 174	–	–	–	Unequal competition (sulfonates)	Vijayaraghavan <i>et al.</i> 2008c
Rx	RK5	B	<i>Corynebact glutamicum</i>	H	HNO <sub>3</sub>	1-3	(25)	L	169-185	2 <sup>nd</sup>	–	En	Electrostat: low pH; Chemical: high pH	Won <i>et al.</i> 2006
Rx	RB4, RO16, RY2	B	<i>Corynebact glutamicum</i>	H	HNO <sub>3</sub>	2	–	L	155-185	–	–	–	Electrostat: low pH; Chemical: high pH	Won <i>et al.</i> 2008
Rx	RY2	B	<i>Corynebact glutamicum</i>	H	HNO <sub>3</sub>	1-4	(25)	L	154-178	–	–	–	Regenerate by raising pH.	Won & Yun 2008
Rx	RO16	B	<i>Corynebact glutamicum</i>	H	HNO <sub>3</sub>	2	(25)	L	157	–	FTIR bonds	–	Electrostat: low pH; Chemical: high pH	Won <i>et al.</i> 2009b
Rx	RO113		<i>Phaneroch. chrysosp.</i>	–, A	–	4		F	19	–	–	–	–	Yalcin <i>et al.</i> 2010
Rx	RR24	Ag	Mod. wheat residue	–	–	4.6	(26)	–	200	–	–	–	–	Zhong <i>et al.</i> 2011

Dye Class	Dye	Biomass class	Biomass	Dried, never, heat	Modification	Best pH	Best temperature	Isotherm best fit	Adsorp. capac. (mg/g)	Rate law best fit	Functional groups	Thermodynamics	Key Findings	Author (year)
Vt	Indigo carmin.	AC	Apricot stone AC	–	H <sub>3</sub> PO <sub>4</sub> , KOH	–	70	L, F	552	–	–	En	–	Gad & Daifull. 2007
Sr	SK	F	<i>Aspergillus n.</i>	N	Homog.	–	(30)	–	–	–	–	–	Fungi tolerant of 10-1000 ppm of dye.	Ali <i>et al.</i> 2008
Sr	SK	F	<i>Aspergillus niger</i>	N	Homog.	(neut)	(30)	–	–	–	–	–	Shaking enhanced uptake.	Ali <i>et al.</i> 2008
Ds	DO30	AC	Carbox bl.	–	Phos. acid	–	–	L	133	–	–	–	Optimization of activation conditions	Ahmad <i>et al.</i> 2009b
Ds	Amino benz.	Ce	Viscose rayon	–	Deriva-tives		20-90	–	0.1-3	–	–	–	Dispersion forces	Aspland & Bird 1961
Ds	DR1	F	<i>Aspergillus niger</i>	A	NaOH ground	(4)	–	–	0.2-6	–	–	–	Functional groups control adsorption	Fu & Viraraghavan 2002a
Ds	DR1	F	<i>Aspergillus niger</i>	A	NaOH, other	4	–	RP	–	–	–	–	Moderately effective	Fu & Viraraghavan 2011
Ds	DO25	AC	Act. C from <i>E. rigida</i>	–	H <sub>2</sub> SO <sub>4</sub> activat.	4-9	20	L	80-119	2 <sup>nd</sup>	–	En	No pH effect.	Gercel <i>et al.</i> 2008
Ds	Bril R, Blue	Ag	Bark, rice husk, cotton	H	–	–	(30)		0-120, 0	–	–	–	Bark > cotton > rice husk	McKay <i>et al.</i> 1986

Dye Class	Dye	Biomass class	Biomass	Dried, never, heat	Modification	Best pH	Best temperature	Isotherm best fit	Adsorp. capac. (mg/g)	Rate law best fit	Functional groups	Thermodynamics	Key Findings	Author (year)
Nk	TY	Al	<i>Chlorella v. algae</i>	N	Living	–	–	–	N.A.	–	–	–	Bioconversion & COD removal	Acuner & Dilek 2003
Nk	O2	F	<i>Aspergillus n.</i>	N	Homog.	–	(30)	–	–	–	–	–	Fungi tolerant of 10-1000 ppm of dye.	Ali <i>et al.</i> 2008
Nk	Drimar-lne B	F	<i>Aspergillus n.</i>	N	Homog.	–	(30)	–	–	–	–	–	Fungi tolerant of 10-1000 ppm of dye.	Ali <i>et al.</i> 2008
Nk	Gris Lanaset	F	Fungal pellets	N	Shaken	(4.5)	(25)	L	70	–	–	–	Adsorption, then fungal degradation; diffusion	Aretxaga <i>et al.</i> 2001
Nk	9 dyes	F	Fungal pellets	D	Pestle, blender	High	30	–	–	–	–	–	Stirring helped; dead biomass was effective.	Asma <i>et al.</i> 2006
Nk	L.green SF (yel)	FA Ag	Bottom ash; deoiled soy	H	Ash heat to 500C	7	50	L	12-25; 8-13	2 <sup>nd</sup>	–	En	–	Mittal <i>et al.</i> 2010
Nk	Food dyes	AC	Charred MCC	–	Torrefac-tion	–	–	–	3-5	–	–	–	Charring essential; NaOH regeneration	Tabara <i>et al.</i> 2011



**Notes for Table A****Dye classes** (in order of appearance):

Ba = basic (positively charged)

Ac = acid (negatively charged, highly soluble)

Dir = direct dye (negatively charged, long or large molecules, substantive)

Rx = reactive dye, usually having sulfonate groups to confer water-solubility

Vt = vat dye, reduced to cationic form for solubilization, then oxidized to insolubilize on the fibers

Sr = sulfur dye, reduced to cationic form for solubilization, then oxidized to insolubilize on the fibers

Ds = disperse dye, inherently hydrophobic, but stabilized by co-additives to allow application

**Dyes:**

AB = Acid Blue

AcO = Acridine Orange (a basic dye)

AO = Acid Orange

AR = Acid Red

AY = Acid Yellow

AmK = Amido black, an acid dye

BPhB = Bromophenol blue (acid dye)

BG = Brilliant Green, a basic dye closely related to Malachite Green

CR = Congo Red, an acid dye, sodium salt of benzidinediazo-bis-1-naphthylamine-4-sulfonic acid

CV = Crystal Violet (also Methyl Violet 10 B), a basic dye, hexamethyl pararosaniline chloride

DB = Direct Blue

DG = Direct Green

DR = Direct Red

DV = Direct Violet

DW = Direct Brown

IC = Indigo carmine, an acid and pH indicator; below pH 11.4 it is blue; above pH 13 it is yellow

MB = Methylene Blue, a basic dye,  $C_{16}H_{18}N_3SCl$ , 3,7-bis(dimethylamino)-phenothiazine -5-ium chloride

MG = Malachite Green, a basic dye, 4-[(4-dimethylaminophenyl)phenyl-methyl]-N,N-dimethylaniline

MO = Methyl Orange, an acid dye, 4-dimethylaminoazobenzene-4'-sulfonic acid sodium salt

MR = Methyl Red, a pH indicator dye; listed here with the basic dyes

MV = Methyl Violet, a group of three basic dyes, MV2B, MV6B, and MV10B

PR = Phenol Red (acid dye)

RB = Reactive Blue

RO = Reactive Orange

RR = Reactive Red

RY = Reactive Yellow

RhB = Rhodamine Blue; [9-(2-carboxyphenyl)-6-diethylamino-3-xanthenylidene]-diethylammonium chloride

SK = Sulfur Black, insolubilized by oxidation

TB = Toluidine Blue-O (a basic dye often used for analysis of colloidal charge)

TY = Tectilon Yellow 2g, an azo dye with amine and sulfonate functionalities

VB = Victoria Blue

Var = Various (in a mixture)

Calmagite, an acid dye, 3-Hydroxy-4-[(2-hydroxy-5-methylphenyl)azo]-1-naphthalenesulfonic acid

**Biomass Classes**

AC = Activated carbon  
Ag = Agricultural residue  
Al = Algae  
B = Bacteria  
Ce = Cellulose (e.g. regenerated cellulose)  
F = Fungal biomass, including yeast  
FA = Fly ash (burned in presence of oxygen)  
Ma = Marine plants (live in the water)  
N = Nuts, including pits and shells  
Pe = Peat  
Pl = Plants, other than agricultural, marine, or wood  
Sl = Sludge  
Tx = Textile materials, including cotton and jute, etc.  
W = Wood, including pinecones and bark

**Drying**

L = living (a subclass of “never dried”)  
N = never dried  
D = dried (e.g. room temperature)  
H = heated (above 50 °C) to dry  
A = autoclaved

**Isotherms**

F = Freundlich  
J = Jossens  
L = Langmuir  
LF = Langmuir-Freundlich  
RP = Redlich-Peterson  
T = Tempkin