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Review Article
Toxicity of Disperse Dyes and its Removal from Wastewater Using Various Adsorbents: A Review

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Abstract

Discharge of colored effluents into surface water bodies pose adverse effects to aquatic ecology and human beings due to mutagenic and carcinogenic effects of dyes responsible for producing color. Disperse dyes cause risk of sensitization and elicitation to the body when colored clothes contact to the body. It can also be regarded as potential allergens to children with suspected contact sensitization. This study enlisted various dyes have been compiled for the adsorption to provide a detailed up summary of available information on a variety of adsorbents such as activated carbon, CFA, orange peel etc. The adsorption capacity of adsorbents are increased by modifying the surface area in various ways and also varying different parameters like as adsorbent dose, adsorbate concentration, agitation speed, temperature, pH, particle size and contact time to minimize various contaminants present in the industrial wastewater. The role of adsorption isotherms and kinetics through various models are the factors influencing the adsorption capacity.

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INTRODUCTION

In this modern society, because of rapidly changing in style and fashion, clothes and colorful products, the usage of dyes increase. According to Confederation of Indian Textile Industry, textile mills are discharging approximately 1.2×10^3 million liters per day (MLD) of colored wastewater into the natural water bodies without proper treatment[1]. There are 3441 textile mills in India out of which 3244 are spinning and 197 are composite mills. The annual consumption of synthetic and natural fibers/filaments is 6800 and 2601 million kilograms respectively. Total production of Khadi (handloom) and other cloth materials is 62625 million kilograms. India's export business of textiles and clothing including silk, jute, coir and handicrafts is about \$300 billion (2013-2014)[2].

In textile mills, there are two principal processes that contribute to the release of dyes in the environment: The cleaning of the dye tank following the preparation of the dye bath and draining of the dye bath after the dyeing process is complete[2]. During a typical dyeing process, the dye bath is prepared, which involves the addition of the dye and a dispersing agent to the dye bath[2]. The draining of the dye bath following the dyeing process that contains the remaining dye which has not been absorbed onto the fiber contributes to the most significant environmental release of dyes, accounting for approximately 90% of the total environmental release of dyes from textile mills.

The release of effluents without proper treatment into water stream, such as river, lakes, etc., pose adverse effects to aquatic life as well as on humans beings. Dye consuming industries like textile mills use large volumes of water and chemicals (dyes) for processing of spins. Approximately 8-20% unutilized dyes and auxiliary chemicals are discharged into the wastewater stream from textile dyeing operations[3]. Approximately 1-10% of pigments used in paper and leather industries are lost as waste. Only half of the dyes can be reduced through effluent treatment processes for colored wastewater. Thus, about hundreds of tons of dyes are discharged daily into the environment as aquatic waste[4].

Dyes: Presently, more than 1.0×10^5 commercially available dyes are in the world whose production was more than 7.0×10^5 t annually[5]. Dyes may be classified in several ways based on their commercial names and chemical nature. Dyes are complex unsaturated aromatic compounds fulfilling characteristics like intense color, solubility, substantiveness and fastness. Dyes can be defined as the different type of coloring particles which differ in each type from the other in chemical composition and are used for coloring fabrics in different colors and shades. Some examples of different dyes are solvent, reactive, acid, paper, plastic, wool, metal complex dye and salt free dyes.

Classification of dyes: Although dyes can be classified in different ways, their primary classification is based on the fibers they are used to color and the chemical composition/properties of different dyes. Dyes can have different chemical nature like water soluble acidic and basic dyes, complex disperse, azo and anthraquinone based dyes together with metal complex dyes[6]. Acid dyes, basic and direct dyes have water soluble components but disperse dye that is less soluble, is very useful for dyeing polyester fibers[6].

Dyes for polyamides and proteins: Polyamide and proteins such as wool, silk and leather have cationic charge on surface therefore dyes used to color such fabrics should contain reverse charge for high electrostatic interaction between fibers and dye molecules. Anionic dyes also known as acid dyes such as acid black 1 can be utilized for coloration only if they are applied to a suitable substrate from an acidic medium but the utilization of these dyes for polyester, cellulosic, or cationic fabrics is highly limited as these polymers, cannot form an ionic bond with anionic dyes[7].

Dyes for anionic polymers: In this type electrostatic force is mainly responsible for dyeing process. Anionic polymers carry a negative charge in their backbone thus; cationic dyes (Dyes bearing a positive charge on their chromophore) are used, for coloring such polymers resulting in a strong ionic interaction between dye and polymer. Also known as basic dyes, cationic dyes are usually water soluble (for e.g., basic red 18) and can react with the anionic sites on the surface of the substrate. The basic dyes produce brilliant colors due to their high tinctorial value[8].

Dyes for cellulosic polymers: Hydrophilic nature of cellulosic polymers like cotton, rayon, cellophane, linen and paper necessitates the use of water soluble (hydrophilic) dyes for their coloration. Direct, azoic, vat, sulfur and reactive dyes can be used for dyeing cellulosic polymers. The name 'Direct dye' alludes to the fact that these dyes do not require any form of 'Fixing'. They are the first known colorants that were utilized for coloring cotton fabrics in the absence of a binding agent known as a mordant[9]. These dyes mostly have low wet fastness due to their hydrophilic nature and are able to gain close proximity to the cellulose chain, resulting in optimization of intermolecular interactions, such as hydrogen bonding[10].

Vat dyes are a large class of insoluble dyes that are classified as such due to the vatting process associated with their application, while sulphur dyes are so named because they contain sulphur as an integral part of the chromospheres as well as poly-sulphide side chains. Both vat and sulfur dyes are insoluble dyes and have no affinity for cellulosic fibers as such but they are converted to a water soluble form by treatment with a weak alkaline solution of sodium sulphide or any other reducing agent to form a leuco compound that is not only water soluble but also has an affinity for cellulosic material[11].

These are subsequently converted back to their water insoluble form, giving good stability under wet conditions. Anthraquinone and indigoids are the two most commonly used vat dyes. Last in the list of suitable colorants for cellulosic fibers is a class of highly colored organic substances called as reactive dyes, that attach themselves to their substrates by a chemical reaction that forms a covalent bond between the molecule of dye and that of the fiber. These are the reactive dyes that made it possible to obtain bright wet-fast shades on cellulosic fibers that were not previously attainable[12].

Dyes for polyesters: Imparting color to polyester is extremely difficult because it is a synthetic fabric made from petroleum, due to which it lacks ionic properties and is essentially hydrophobic in nature. Dyeing polyester thus necessarily requires the use of disperse dyes. Other types of dyes leave the color of polyester almost entirely unchanged. Disperse dyes are a class of sparingly soluble synthetic organic dyes with free ionizing group that have no affinity for hydrophilic polymers such as cellulose, cellophane and paper, but are quite suitable for poly ethylene terephthalate and cellulose acetate[13]. Disperse dyes are applied in acidic condition at high temperature. Due to lesser cost, a wide range of colors and better dyeing properties they offer quick replacement of traditional natural dyes. Among various dyes, disperse dyes are commonly used in pulp and paper mill, textile mill and plastic industry in India. The solubility of disperse dyes in water is very less due to their crystalline structure with varying particle size. Disperse dyes are scanty in dispersing and cause unbalancing composition of dye during dying process[14]. Generally, dispersing media are anionic, poly-condensates of aryl-sulphonic acids or lignin sulphonates with formaldehyde, which assists grating by thwarting agglomeration of the dye particles. Disperse dyes are preferred over acrylic black O, red GTL and others dyes due to their high tendency to bind the fibers and the fact that these remain persistent over the years. However, these dyes become toxic in the water bodies due to their complex molecular structure with fused aromatic groups[15].

Ions of disperse dye in water streams either reflect back the solar radiation or scatter in the water bodies. Solar radiation cannot penetrate beyond littoral zone and therefore, the deep water zone (i.e., aphotic zone) develops anaerobic condition. The entire ecological cycle including self-purification system of water stream is disturbed due to lack of dissolved oxygen[16]. The indiscriminate discharge of untreated wastewater is more conspicuous in case of developing countries rather than developed one, where the shortage of modern technology become advanced knowledge and insufficient funds are aggravating the problems[17].

Disperse dyes have different chemical groups in various percentage such as mono azo dyes (50%), anthraquinonoid dyes (25%), diazo dyes (10%), methyne dyes (03%), styryl dyes (03%), acrylene benzimidazol (03%), quinophthalone dyes (03%), amino naphthyl amide (01%) and naphthoquinone imine (01%). Azo dyes are characterized as the largest group of disperse dye for two reasons: One having large number of molecular combinations generated, which results in large variety of dyes and other having a simple, easy and low cost of manufacturing process as compared to the expensive anthraquinone dyes[2]. By virtue of their simplicity and the ease of production, the manufacturers can respond much more easily to customer's and end user's requests for special shades and fastness characteristics having low cost[18]. Disperse blue 79:1 (DB) and disperse orange 25 (DO) dyes as having azo group are toxic which enter in the environment in such quantities or concentrations that have direct or enduring harmful effect on the environment as well as the aquatic system by producing metabolites during their formulation.

In this study, the technical feasibility of various low-cost adsorbents for dye removal from colored wastewater has been assessed. The main objective of this study was to provide a summary of recent information concerning the use of low-cost materials as sorbents. For this, an extensive list of sorbent literature has been compiled. This study described (i) Toxicological effects of disperse dyes (ii) A critical analysis of these materials and (iii) Various mechanisms involved during the process.
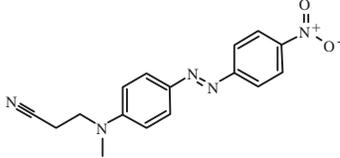
Toxicity due to disperse dyes

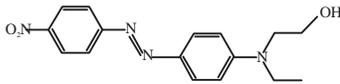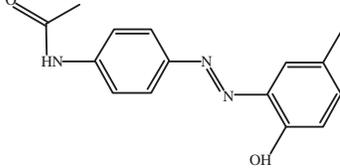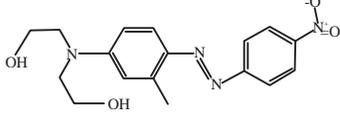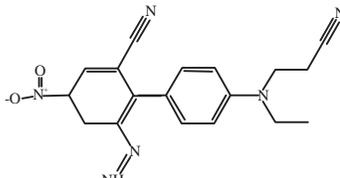**Allergies from dyes:** Contact allergic reactions to synthetic textile dyes have been noticed since 1868[19,20]. In 1884, due to the influence of new synthetic dyes in the market, the numerous complaints were seen as unacceptable skin eruptions, caused by wearing, when in the state of

perspiration hosiery and flannel colored with aniline dye's[21]. In 1940, nylon stockings were introduced into the American market. Soon after their introduction, cases of nylon stocking dermatitis were reported. This was originally called 'nylon allergy'. Upon further investigation, however, it was found that the dermatitis was caused by dyes and not by nylon[22,23]. Dobkevitch and Baer[24] reported eczema due to cross hypersensitivity between the azo dye disperse yellow 3 in nylon stockings and the para-amino compound p-phenyl enediamine, today mostly used as a hair dye. In the beginning of 1985, patients were found with Allergic Contact Dermatitis (ACD) disease resulted from dark clothes, mostly because of contact allergy to other disperse dyes, such as disperse blue[25-27] 106 and disperse blue 124.

Aquatic toxicity: Colored substances present in aquatic living organisms have been associated with changes in protozoan colonization rate, phytoplankton species composition and primary productivity. In addition, secondary production, macro-invertebrate behavior and macro-invertebrate community system can be affected. Color can also alter the availability and toxicity of heavy metals to fishes[28]. Apart all aquatic bodies poses severe environmental problems because dyes effect the toxicity to the aquatic life, affect self purification system and are damaging the aesthetic nature of the environment. Most of the industrial use dyes are stable to light and oxidation. Use of dyes and pigments in the effluents cause collective toxicity which affects the aquatic ecosystem and aesthetic nature of the surroundings[29]. The discharge of industrial effluents containing toxic components which enter the surface water system, stimulate much attention due to their potential to cause the environmental problem (Table 1). These pollutants increase the adverse consequences in the color, odor and taste of surface water[30].

Table 1: Disperse dyes and their toxicity

Disperse dye name	Structure	Toxicity
Disperse orange 25		Asthma-like symptoms, not readily biodegradable, Development of bladder cancer
Disperse blue 79:1		Mutagenic activity, not readily biodegradable, Asthma-like symptoms
Disperse red 1		Mutagenic activity, cyanosis, development of bladder cancer
Disperse yellow 3		International agency for research on cancer (IARC): Group 3
Disperse red 17		Skin irritation, mutagenicity/genotoxicity, one generation reproduction toxicity
Disperse red 73		Skin irritation, eye irritation
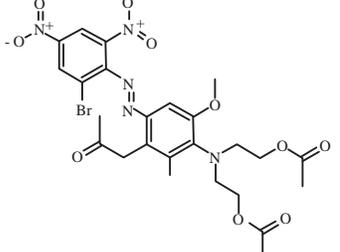
Dyes removal techniques: Number of techniques are available for decolorization of wastewater like Fenton process[31], ozonation[32], membrane separation[33], coagulation/flocculation[34], electro-coagulation and flocculation[35], co-precipitation[35], oxidation[36], ion exchange resin[37], membrane techniques like ultra-filtration[38], nano-filtration[39], reverse osmosis[40], electrolysis[36], degradation by micro-organisms, photochemical[41] and adsorption[36] using different kinds of adsorbents.

Fenton process: Off lately there has been a growing demand for utilization of advanced oxidation processes (AOPs) as an attractive alternative for decolorization and degradation of dyes as well as for the destruction of a large number of persistent organic pollutants. Among the list of various available AOPs, Fenton reagent (H_2O_2/Fe^{2+}) has proved to be a promising tool for organic pollutant oxidation. Particularly in Fenton process H_2O_2 is used in the presence of $FeSO_4$, to decolorize the effluents at a higher rate and at higher concentrations of the reagents. Ince and Gonenc[42] demonstrated degradation of the dye intermediate anthraquinone-2-sulfonic acid sodium salt by using Iron (III) with hydrogen. Ince and Gonenc[42] used UV/H_2O_2 for oxidation process for the decolorization of reactive black 5 in textile effluent. The degradation of anthraquinone sulfonate dyes have been analyzed by Kiwi *et al.*[43] in presence of H_2O_2/TiO_2. In yet another study done by Morrison *et al.*[44], it was observed that the pH of the solution had immense bearing on the rate of decomposition when a non-biodegradable azo dye orange II was effectively mineralized with iron and hydrogen peroxide.

The advantage of the Fenton reagent is that no activation energy is required for activating hydrogen peroxide[45]. Thus, this process offers a cost-effective method of generating hydroxyl radicals, utilizing easy-to-handle reagents. But a major disadvantage of this method is the production of a substantial amount of $Fe(OH)_3$ precipitate and non retention of the homogeneous catalyst added as an iron salt which further contributes to additional water pollution.

Ozonation: Ozonation is another process which is fundamentally an oxidation process. Ozone is an unstable gas which can degrade back to oxygen and during this transition a free oxygen atom, or free radical is formed. The free oxygen radical is highly reactive to disinfection of bacteria or organic moiety. Over the last decade, bubbling induced ozonation or catalytic ozonation has been widely employed in large scale for water and wastewater treatment. Hao *et al.*[46] employed ozonation in the decolorization of orange II containing water whereas oxalate, formate and benzene sulfonate ions were the most important decomposition products.

There are some advantages of ozonation like at an acidic pH the rate of decomposition is considerably high. Ozone rapidly reacts with substrate and does not require additional chemicals to water. As observed by Cuerda-Correa *et al.*[47], there is negligible influence of temperature and UV irradiation on the rate of decomposition. But, on other hand higher equipment and operational cost does not support its user friendly utilization. Besides, ozonation by-products can create possible hazardous effects on health and sparingly soluble nature of ozone requires special mixing techniques.

Membrane separation: In membrane separation, a semi-permeable phase controls the rate of movement of various molecules between the phases. Membrane and ozonation process was used together for treatment of reactive dye containing wastewater[48]. Bamperng *et al.*[49] used ozonation membrane contacting process for direct red 23, acid blue 113 and reactive red 120. Membrane process usually does not require phase changes thus less amount of energy is needed to proceed. Membrane separation is eco friendly and deals with simple and harmless materials. Some drawbacks can also be associated with membrane separation as it cannot be easily staged and fails to operate at above room temperature. Membrane processes often do not scale up to accept massive stream sizes and also have chemical incompatibilities[50].

Chemical oxidation: A recent addition in the list of available methods for treatment of colored effluents is the chemical oxidation of dyes by utilizing ultraviolet light (UV) and a strong oxidant[51]. The most commonly utilized oxidant for this purpose is hydrogen peroxide (H_2O_2) used in combination with UV. Several studies have already established the oxidative capabilities of chlorine dioxide (ClO_2) and its potential as an effective decolorant. This has facilitated the usage of UV in combination with ClO_2 for the reduction of colored effluent generated from textile industries[52].

Photochemical degradation: Dye degradation in presence of H_2O_2, Fenton process and ozonolysis has successfully eliminated color from effluents. Now-a-days, nano science has started stepping into remediation process due to large surface area over bulk[53]. Liu *et al.*[54] used UV irradiation and titanium dioxide (TiO_2) catalysis for the decomposition of azo dye and demonstrated that TiO_2 has photocatalytic efficiency for azo dye acid black 1 in water. Bin and Sobera-Madej[55] studied the photocatalytic decomposition of textile dyes on TiO_2-titania

A11 and TiO_2-degussa P25. Rahman *et al.*[56] could conclude from their observations that photo catalytic degradation of dyes often results in formation of by-products during the process of degradation.

Microbiological decomposition of synthetic dyes: The use of microorganisms for the removal of synthetic dyes from effluents offers considerable advantages because of relative inexpensiveness and generation of non toxic end products. Kanekar and Sarnaik[57] developed an activated sludge process utilizing strains of microorganisms derived from bovine fecal waste for the removal of methyl violet and rhodamine B. But the stable chemical composition of the majority of such compounds makes them highly resistant to microbiological attack. To add to this, a well furnished lab and trained handling is required for bacterial culture and their application.

Coagulation/flocculation: Coagulation/flocculation process is widely used, simple and cost-effective process. The coagulation/flocculation are combination of two distinct procedures, the first one is coagulation, in which destabilization of a given colloidal suspension or solution takes place[34]. Coagulation helps to overcome the factors that promote the stability of a given system. This is achieved by utilizing suitable coagulating agents like aluminium or iron salts. Coagulation is followed by flocculation; a process which facilitates the induction of destabilized particles so that they come in contact with each other and thereby, form large agglomerates, which can easily be separated through methods like gravity settling[58]. The first process i.e., coagulation is completed in a very short duration whereas flocculation requires more time to complete. Electro coagulation is combined technique in which aluminum or iron is used as anode and the coagulating metal cations are released *in situ* as long as an electrical current is applied. Some examples of organic and natural flocculants are acrylamide, acrylic acid, diallyldimethyl ammonium chloride, styrene sulphonic starch, cellulose and natural gums. The treatment of textile dye waste with magnesium carbonate was investigated by Judkins *et al.*[59] and Aad *et al.*[60]. Inorganic coagulants such as alum, magnesium carbonate, vermiculite and ferrous sulfate are widely used for chemical coagulation of dyes. Fast mixing and optimization of pH are the reasons responsible for the limited use of this process. Flocculation demands a continuous attention on the mixing speed and amount of mix energy is predominately dependent upon the type and design of the equipment.

Adsorption: Adsorption is a promising process of decolorization with attachment of dye molecules on surface of adsorbent by physical or chemical interactions[61]. The solid substance on the surface of which adsorption occurs is known as adsorbent. The substances that get adsorbed on the solid surface due to intermolecular attractions are called adsorbate. The process of removal of adsorbed substance from the surface on which it is adsorbed is called desorption. It is the reverse of adsorption and can be brought about by heating or by reducing the pressure. Adsorption is quite different from another similar sounding process called absorption in which substances diffuse into some bulk phase-solid, liquid or gaseous[62]. Adsorption, as a process has wide industrial applications and is even operative in many physical, chemical, biological and natural systems. It is a surface phenomenon and occurs due to the unbalanced forces on the surface of the solids and liquids. If we consider a solid, it is examined that a molecule present in the bulk of the solid is being uniformly engrossed from all sides by the neighboring molecules[63]. As a result, there is no net attraction on this molecule. However, a molecule which lies near the surface (known as a surface molecule) is attracted only by molecules below it. Therefore, surface molecules experience a resultant downward attractive force within the solid or liquid[64]. As a result, the surface of solid or liquid tends to satisfy their residual forces by attracting and retaining the molecules of other species (a gas or dissolved substance) when brought in contact with them. Thus the phenomenon of higher concentration of molecular species (gases or liquids) on the surface of a solid than in the bulk is called adsorption[65] (Table 2).

Various researcher primed starch-based polymers by a cross linking reaction of starch-enriched flour using epichlorohydrin as a cross linking agent in the presence of NH_4OH. The yield, mobility and structural properties of cross linked starch materials with various compositions were investigated and found a correlation between the structure, mobility and degree of cross linking of these sorbents. The cross linked starch-based materials, containing tertiary amine groups were used for the recovery of various dyes from wastewater. It is recommended that the sorption mechanism was correlated to the structure of the polymer.

One of the most important characteristics of an adsorbent is the quantity of solute it can be accumulated which was usually calculated from the adsorption isotherms. Dyes that are difficult to breakdown biologically, can often be removed by using the various adsorbent. A good adsorbent should generally possess a porous structure which resulting in high surface area and the time taken for adsorption equilibrium to be established should be as small as possible so that it can be used to remove dye wastes in lesser time.

Some of the adsorbents, which are generally used for dye removal from wastewater are: Alumina, a synthetic

Table 2: Multi-component adsorption studies of dyes systems

Adsorbents	Dyes removed	Models and methods	References
Activated carbon	Basic blue 41, basic red 18, remazol reactive yellow, remazol reactive black, remazol reactive red, acid blue 80, acid red 114, acid yellow 117, basic blue 69, basic red 22, basic yellow 21, methylene blue, rhodamine B, atrazine and congo red	Langmuir, Freundlich	Noroozi et al.[66], Porter et al.[67], Choy et al.[68], Valix et al.[69], McKay and Al Duri[70], McKay and Al Duri[71], McKay and Al Duri[72] and Pelekani and Snoeyink[73]
Silkworm pupa	Basic red 18	Langmuir	Noroozi et al.[66]
Natural zeolite	Malachite green	Freundlich, Langmuir	Wang and Wang[74]
Dried sugar beet pulp	Gemazol turquoise blue-G reactive dye	Langmuir	Aksu and Isoglu[75]
Cross-linked chitosan	Metanil yellow, reactive blue 15	Langmuir	Chiou and Chuang[17]
Lignite	Basic blue 3, basic yellow 21, basic red 22	Ideal adsorbed solute theory (IAST)	Choy et al.[68]
Natural sediment	Atrazine, congo red, methylene blue	Freundlich	Tao and Tang[76]
Peat	Basic yellow 21, basic red 22, basic blue 3	Freundlich, Langmuir, Redlich-Peterson	Allen et al.[77] and Allen et al.[4]
Humic acid, tamarind fruit shell powder	Methylene blue	Freundlich, Langmuir	Vinod and Anirudhan[78]
Almond tree and teak tree bark powder	Crystal violet, rhodamine B	Freundlich, Langmuir	Patil and Kulkarni[79]
Mango and mangrove plant leaf powder	Crystal violet, rhodamine B	Freundlich, Langmuir	Patil et al.[80]
Rhizopus arrhizus biomass	Reactive blue 19, reactive orange 16	Freundlich, Langmuir	O'Mahony et al.[81]
Cellulose membrane	Direct yellow 12, direct blue 15	Diffusion	Maekawa et al.[82]
Bentonite	Basic yellow 28, basic red 46	Freundlich, Langmuir	Turabik[83]
Husk of mango seed	Acid blue 80, acid blue 324, acid green 25, acid green 27, acid orange 7	Freundlich, Langmuir	Davila-Jimenez et al.[84]
Rarasaponin-bentonite	Malachite green, methylene blue	Extended Langmuir model	Kurniawan et al.[85]
Palm Kernels and its activation with KOH	Acid blue 74, basic brown 1 and Methylene blue	Countercurrent two and three-stage	Tseng et al.[86] and Tseng et al.[87]
Activated clay, montmorillonite, Activated carbon	Basic violet 10, basic green 5	Freundlich, Langmuir, Thermodynamic parameters	Shiau and Pan[88]
Carbon slurry	Vertigo blue 49 and orange DNA13	Langmuir isotherm, thermodynamic	Gupta et al.[89]
Char-fines, lignite coal, bituminous coal and comparing results with activated carbon (Filtrasorb-400)	Trisazo direct dye, direct brown 1:1	Langmuir adsorption model	Mohan et al.[90]
Bacterial-yeast consortium BL-GG	Scarlet RR	Veratryl alcohol oxidase, laccase, tyrosinase and NADH-DCIP reductase	Kurade et al.[91]
Orange peel, banana Peel , neem leaves	Methyl orange, methylene blue, rhodamine B, Congo red, methyl violet, amido black 10B	Batch wise column	Velmurugan et al.[92]
White marble	Maxilon blue GRL	Langmuir and Freundlich isotherm	AlJebori and Alshirifi[93]
Orange peel	Direct red 23, direct red 80	Langmuir and Freundlich isotherm, Kinetics	Arami et al.[94]
Microcrystalline cellulose	Congo red	Batch adsorption ultasonic pretreatment	Hu et al.[95]
Enterobacter gergoviae	Disperse brown 21	Aerobic condition	Benmerine et al.[96]
Acacia arabica fruit	Methylene blue	Hybrid response surface methodology	Saibaba and King[97]
Montmorillonite clay	Methylene blue	Langmuir and Freundlich isotherm, kinetics	Almeida et al.[98]
CFA	Disperse blue 79:1 and disperse orange 25	Langmuir and Freundlich isotherm, kinetics	Kisku et al.[99]
Tea leaves	Methylene blue	Langmuir and Freundlich isotherm, kinetics	Shukla et al.[100]
Modified cenospheres	Disperse blue 79:1 and disperse orange 25	Langmuir, Freundlich and temkin isotherm, Kinetics	Tiwari et al.[14]
Sawdust	Methylene blue	Langmuir and Freundlich isotherm, kinetics	Shukla et al.[100]
Cellulose based (Banana and orange peels)	Methyl violet, amido black 10B	Langmuir and Freundlich isotherm	Annadurai et al.[101]

porous crystalline gel, which is available in the form of granules of different sizes having surface area ranging from 200-300 $m^2 g^{-1}$. Bauxite a naturally occurring porous crystalline alumina composition of kaolinite and iron oxides normally having surface area ranging from 25-250 $m^2 g^{-1}$. Alumina has been tested by various workers for the removal of dyes[62]. Silica gel, prepared by the coagulation of colloidal silicic acid resulted in the formation of porous and noncrystalline granules of different sizes. It showed a higher surface area as compared to alumina, which ranges from 250-900 $m^2 g^{-1}$.

Adsorption capacity of adsorbent is defined as mass of solute adsorbed per unit mass of adsorbent and nature of adsorption can be described by relating the adsorption capacity to equilibrium concentration of the solute remaining in the solution using various isotherms[14]. Data were tested for suitability of isotherms such as Langmuir, Freundlich and Temkin. Langmuir isotherm assumes that the adsorption takes place at specific homogeneous sites within the adsorbent. linear form of Langmuir isotherm is presented as Eq. 1:

$$\frac{1}{q_e} = \frac{1}{q_m} + \left(\frac{1}{b\, q_m}\right)\frac{1}{C_e} \tag{1}$$

where, C_e is the concentration of adsorbate in solution at equilibrium (mg L^{-1}), q_e is the amount of dye adsorbed on adsorbent at equilibrium (mg g^{-1}), q_m is the maximum quantity of dye required to form a single monolayer on unit mass of adsorbent and b is a parameter for apparent energy of adsorption, defined as $b = (k_a/k_d)$.

Additionally, isotherm may be expressed in terms of separation factor R_L which is a dimensionless constant and can be determined by the following Eq. 2.

$$R_L = \frac{1}{1 + K_L C_0} \tag{2}$$

where, C_0 is the initial dye concentration (mg L^{-1}), K_L is the Langmuir constant related to the energy of adsorption.

The value to R_L indicates the shape of the isotherms to be either unfavorable ($R_L > 1$), linear ($R_L = 01$), favorable ($0 < R_L < 1$) or an irreversible ($R_L = 0$).

Freundlich isotherm is derived by assuming heterogeneous surface with a non-distribution of heat of adsorption over the surface. The linear form of the Freundlich isotherm is expressed by Eq. 3.

$$\ln q_e = \ln K_f + \left(\frac{1}{n}\right) \times \ln C_e \tag{3}$$

where, C_e is the concentration of adsorbate in solution at equilibrium (mg L^{-1}), q_e is the amount of dye adsorbed on adsorbent at equilibrium (mg g^{-1}), n is the Freundlich exponent constant that represents the parameter characterizing Quasi-Gaussian energetic heterogeneity of the adsorption surface and K_f is the Freundlich constant indicative of the relative adsorption capacity of the adsorbents (L g^{-1}).

Temkin isotherm suggested that the heat of adsorption of all molecules in layer decreases linearly with coverage due to adsorbent-adsorbate interactions and the adsorption is characterized by a uniform distribution of the bonding energies, upto maximum binding energy. Temkin isotherm is represented by Eq. 4:

$$q_e = B_t \ln K_t + B_t \ln C_e \tag{4}$$

where, C_e is the concentration of adsorbate in solution at equilibrium (mg L^{-1}), q_e is the amount of dye adsorbed on adsorbent at equilibrium (mg g^{-1}), K_t is the equilibrium binding constant (L mg^{-1}) and B_t is the variation of adsorption energy (kJ mol^{-1}).

Controlling parameters during adsorption process

Effect of temperature: The adsorption process for dye removal is dependent on an important variable called temperature. Dyes containing industrial effluents have a high temperature in the range of 60-70°C and thus their untreated discharge into water bodies is a major source of thermal pollution. Nigam *et al.*[102] studied the effect of temperature variation from 5-35°C on the removal of direct dyes by *Trametes versicolor* biosorbent and reported an increase in removal efficiency with an increase in temperature. This might be due to rise in the motion of dye molecules. Akhtar *et al.*[103] explored the effect of varying temperature on the biosorption of 2,4-dichlorophenol. When temperature increased from 0-300°C, the percentage sorption was increased by 98%. With a further increase in temperature up to 600°C, the percentage of biosorption was found to decrease. This might be attributed to the alterations in the structure of the biomass. Bulut *et al.*[104] made similar observations while studying the adsorption of Direct blue 71 by wheat shells. The effect of different temperatures (20, 30 and 40°C) on the biosorption capacity was noted and it was found that the adsorption capacity increased with increase in temperature. The process was endothermic in nature.

Temperature has two main effects on the surface adsorption process. Increasing the temperature is thought to accelerate the rate of diffusion of the adsorbate molecules

across the external boundary layer and within the internal pores of the adsorbent particles as results of the reduced viscosity of the solution[69]. Furthermore, changing the temperature, changes the point of equilibrium of the reaction between adsorbent and the selected adsorbate. Studying the dependence of the adsorption processes on the temperature variation has considerable importance regarding the physical property and entropy changes related to surface assimilation[105]. Gundoan *et al.*[106] observed the effect of temperature on the adsorption of MB dye (initially at 700 mg L^{-1}) onto MCasa (MC) as a function of time. The amount of methylene blue adsorbed increased from 245 mg g^{-1} (68% removal) to 300 mg g^{-1} (74% removal) as temperature increased from 35-60°C which also establishes percentage removal as a function of temperature. The increase of 6% in methylene blue dye removal for a 25°C increase in temperature indicated that temperature has a significant role and also that the adsorption of methylene blue dyes onto the MC adsorbent surface was an endothermic process. Upto the point of equilibrium rate of dye removal showed a direct relation with increase in temperature indicating a kinetically controlled process, as found in many other systems[106]. The mobility of molecules increases normally with increase in temperature, thus facilitating the formation of surface monolayer.

The temperature effect on adsorption of dye to Ca(OH)$_2$ treated fly ash was investigated at four different temperature range (35, 40, 45 and 50°C) by Chowdhury and Saha[107]. They found an inverse relation between adsorption of dye and increase in temperature, establishing that adsorption process was unfavorable at higher temperatures. Decreased sorption capacity of the adsorbent with increasing temperature was an indication of the exothermic nature of the adsorption process.

Sharma and Bhattacharyya[108] studied the effect of temperature on the removal of congo red dye by *Azadirachta indica* leaf powder. They used four different temperature ranges (27, 35, 40 and 50°C) and also observed that the adsorption capacity showed an inverse relation with temperature. With an increase in temperature, the adsorption of the dye molecules decreased, as the adsorbate separated from the solid surface to enter the liquid phase thereby decreasing the adsorption capacity.

Effect of pH of the solution: The pH of the aqueous solution has been established as one of the most important factors influencing the adsorption kinetics. It influences not only the degree of ionization of the solute, the surface charge of the adsorbent and dissociation of functional groups on the active sites of the adsorbent, but also dye chemistry in the solution[109].

Chowdhury and Saha[110] investigated the pH dependency of MG adsorption by alkali treated fly ash. The initial pH of the solution was adjusted by using 0.1 N HCl and 0.1 N NaOH after addition of adsorbent. The maximum adsorption capacity was obtained at pH 6.0-9.0 moreover at pH 7.0, the maximum amount of dye adsorbed per unit mass of sorbent was 4.94 mg g^{-1}. The profile of pH indicates that MG is one of the cationic dyes, it adsorbs to the adsorbent surface at higher pH values. The protonation of the dye takes place at low pH which results in low adsorption of dye in acidic medium. The removal of dye is inhibited at low pH values, mainly due to the increased competition between protons and dye molecules for the same binding sites. With an increase in pH of the solution, the dye becomes more deprotonated; thereby increasing the negative charge density on the surface of the adsorbent thus, efficient removal of dye is observed when the pH of the solution is increased[75].

The pH of the solution substantially influences the adsorption of dye molecule due to change in the surface properties of the adsorbent. The solubility of dye is mainly affected by change in pH concentration of the solution. Namasivayam and Kavitha[111] examined the effect of pH on the removal of direct dye (Congo red) by using orange peel as an adsorbent. The maximum removal (76.6%) was observed at pH 5.0 minimum (49%) at pH 12.0. It is concluded that more removal is obtained in acidic pH of the direct dye solution. In another study, Namasivayam and Kavitha[111] utilized activated carbon prepared from coir pith as a suitable adsorbent for the removal of congo red dye. They studied the effect of varying pH on the rate of removal. In highly acidic condition (pH 2.0, dye concentration, 20-40 mg L^{-1}), the removal percentage was about 70%. At pH 4.0, the removal percentage decreases from 70-50% and reached equilibrium at pH 10.0. The researchers explained that the observed effects could be attributed to the electrostatic attraction between positively charged binding sites on the adsorbent and negative charge bearing dye molecules or to a chemical reaction between biomass and dye. At high pH values, the binding sites of the adsorbent become negatively charged which did not allow the adsorption of dye due to repulsion.

Sharma and Bhattacharyya[108] studied the removal of congo red dye by neem leaf powder. Their findings showed that the uptake of the congo red dye increased when the pH of the solution was increased from 4.0-7.0 and after that pH showed almost negligible effect on the rate of removal. They also concluded that maximum interaction between the surface of neem leaf powder and dye molecules was observed near pH 7.0. The effect of pH variation on adsorption of three dyes namely: Amaranth, sunset yellow and fast green FCF by powdered peanut hull was studied by Gong *et al.*[112].

Gupta *et al.*[89] varied the pH of the solution from 3.0-10.0 and conducted experiments on carbon slurry at a fixed dye concentration, i.e., 300 mg L^{-1}. They observed a significantly high electrostatic repulsion existing between the positively charged surfaces of the adsorbent and dye molecules in acidic conditions. This considerably decreased the adsorption capacity of the adsorbent at low pH values, especially at values lower than 6.2. When the pH of the solution was increased from 6.0-7.0, percentage removal increased dramatically as a strong electrostatic attraction developed between positive charge bearing dye molecules and negatively charged binding sites on the surface of the adsorbent. Increasing the pH beyond 7.0 decreased the rate of adsorption.

Effect of adsorbent dose: The methylene blue adsorption at a contact time of 2 h was studied by varying the adsorbent montmorillonite clay (MC) dose from 1.6-4.6 g L^{-1} in a 600 mg L^{-1} methylene blue solution[113]. It is observed that the percentage removal of methylene blue increased with the increasing adsorbent concentration. An increase in concentration of MC facilitated increased adsorption of methylene blue due to the availability of a greater number of possible binding sites resulting from increased surface area of MC. When the adsorbent dose was increased beyond 3 g L^{-1}, there was slight change in the rate of removal of methylene blue and the rate of attaining adsorption equilibrium. Tor *et al.*[114] studied the removal of malachite green by changing the quantity of adsorbent in the test solution from 0.5-3.5 g. The percentage removal increased when the adsorbent dose was increased and equilibrium was attained at the adsorbent dose of 1.0 g. A possible explanation for this increase in adsorption capacity comes from the increase in the number of available adsorption sites that allow the binding of a greater number of dye molecules onto the surface of adsorbent. Since, increasing the adsorbent dose beyond 1.0 g did not influence the adsorption characteristics notably, this value was considered as the optimal value for further studies as the least amount of fly ash corresponding to the highest adsorption is declared as the optimum dose.

Variation in adsorbent dose is an important factor because it explains the removal capabilities of adsorbent for a meticulous dye concentration. The effect of varying the adsorbent dose on the removal of congo red direct dye was evaluated by Namasivayam *et al.*[115]. The results showed that when adsorbent dose was increased, the percent uptake of dye also increased. The optimum dose (0.5 g) of adsorbent corresponded to the maximum (92%) uptake of dye. This can be attributed to the presence of large surface area of the adsorbent for the given mass. Sharma and Bhattacharyya[108] also studied the effect of adsorbent dose on the removal of congo red dye. The dye removal was maximum at low adsorbent dose of neem leaf powder because the low amount of neem leaf powder showed good relations with dye molecules. Namasivayam *et al.*[115] also reported the removal of congo red by coir pith carbon using different adsorbent dose (2-18 g L^{-1}) at different dye concentrations (20, 40, 60 and 80 mg L^{-1}). They also showed that the maximum percentage removal was obtained at a high adsorbent dose. Gong *et al.*[112] reported the effect of adsorbent dose on the uptake of dyes such as amaranth, sunset yellow and fast green from aqueous solution. The rate of dye removal was found to increase with increase in the adsorbent dose. At high dose (10 L^{-1}), the percent uptake of amaranth, sunset yellow and fast green dyes was 98.64, 98.26 and 99.12%, respectively. It was due to the presence of more exchanging sites on the adsorbent surface which facilitated greater removal of dyes. On the other hand, Namasivayam *et al.*[115] also reported removal of congo red by coir pith carbon using various adsorbent doses (100-900 mg/50 mL) at varying dye concentrations (20, 40, 60 and 80 mg/L). In case of high adsorbent dose, the maximum percentage removal was observed.

Effect of adsorbate concentration: The concentration of adsorbate is also an important variable affecting the adsorption process. In general, an increase in the adsorption of dyes with increase in dye concentration is observed. Namasivayam *et al.*[115] showed that when initial dye concentration increased, the dye adsorption per unit mass of the adsorbent also increased. This demonstrates that the increase or decrease of dye adsorption depends on the initial dye concentration. Gong *et al.*[112] also performed a study to show how the initial dye concentration affects the percentage removal of dyes. They observed that the removal percentage decreased from 99.60-70.40% for sunset yellow, 98.70-72.80% for amaranth and 98.20-74.30% for Fast green with the increase in initial dye concentration due to the accumulation of dye ions at higher dye concentration. Such accumulation decreases the total surface area of the adsorbent particles available for adsorption of dyes.

Mall *et al.*[116] studied the uptake of congo red dye onto bagasse fly ash and activated carbon to see the effect of initial dye concentration. They observed an increase in the amount of dye adsorbed with the increasing initial dye concentration due to increase in the driving forces to overcome mass transfer resistance of the dye between the aquatic and solid phases.

However the study also concluded that the percentage removal of congo red dye decreased with the increase in dye concentration. The effect of varying the initial concentration of 2,4-dichlorophenol onto rice husk was illustrated by Akhtar *et al.*[103]. The initial dye concentration was increased from $0.61\text{-}6.10 \times 10^{-4}$ mol dm^{-3}. Authors recommended that the distribution coefficient decreases with the increase in concentration of dye up to 4×10^{-4} mol dm^{-3} and remains constant thereafter. This might be attributed to the smaller number of binding sites available for the adsorption of a huge number of dye molecules.

Bulut *et al.*[104] also reported similar findings and stated that an increase in initial dye concentration from 50-250 mg L^{-1} increased the amount of dye adsorbed per unit mass of adsorbent. They supported their observations explaining that the binding sites on the surface of the adsorbent remain unsaturated during the initial stages of the adsorption process. The effect of initial dye concentration on the removal of direct red 80 by almond shells was investigated by Ardejani *et al.*[117]. It is showed that the percentage removal of direct red 80 dye decreased from 94.00-83.90% with increase in initial dye concentration from 50-150 mg L^{-1}. Jain and Sikarwar[118] also concluded that amount of dye adsorbed per unit mass of the biomass increased with increase in the dye concentration. The maximum percentage removal (nearly 99%) was observed at the lowest dye concentration (1.0×10^{-5} M) for activated carbon biomass and (nearly 80%) for sawdust. However, the percentage removal was higher at low initial dye concentration, indicating that the initial dye concentration significantly affected the adsorption of direct yellow 12 dye. In yet another study different concentration of direct brown and direct brown 2 dyes were used[85] to analyze the effect of initial dye concentration on the amount of dye adsorbed. They observed that with the increase in the initial dye concentration, the amount of dye adsorbed also increased. Tor *et al.*[113] studied the effect of methylene blue dye concentration on its adsorption onto MC and the percentage dye removal. It is proposed that the initial dye concentration significantly affected the adsorption potential of MC. At maximum solution concentrations, viz., 1400 and 1500 mg L^{-1}, the adsorption of methylene blue showed a minor decrease. These changes in equilibrium indicate the occurrence of a chemisorptions process at the surface of the MC in the initial stages of the experiment, followed by a second adsorbate layer in the later stages, whereas the adsorption capacity decreases in the final stage, thus resulting in dimerization of methylene blue dye at high concentrations. The ionic nature of methylene blue dye could be responsible for the observed behavior, leading to one or more reactions over and above the primary adsorption phenomena[119].

Chowdhury and Saha[110] found that absorption potential of treated fly ash was a function of dye concentration. The sudden profile of the curves obtained at the beginning of the experiment showed that the rate of adsorption was high, suggesting the appropriateness of Ca (OH)$_2$ treated fly ash for the treatment of very dilute dye solutions. It was also observed that the rate of adsorption increased quickly in the starting and became slow in the end. Initial faster rates of adsorption were due to the availability of more binding sites for adsorption and the slower rates of adsorption at the end were due to the saturation of the binding sites and attainment of equilibrium. It was observed that when the initial dye concentration was increased from 10-100 mg L^{-1}, the adsorption potential of the adsorbent increases due to the presence of a driving force that helps to conquer all the mass transfer resistances of dyes between the aqueous and solid phase. However, the percentage of sorption decreases with the increase in initial dye concentration. It is explained that the limited number of active sites on the adsorbent become saturated at a definite concentration. This establishes that the adsorption capacity is bound to increase with the increase in initial dye concentration mainly due to the rise in mass transfer from the concentration gradient.

Effect of agitation speed: The strong dependence of adsorption process on agitation speed emanates from the facilitated mixing of solute in the solvent and the formation of the external boundary film. Shiau and Pan[88] studied the effect of agitation speed on film thickness for adsorption process. They conducted a series of experiments at various agitation rates (200-600 rpm) to study the adsorption of Basic green 5 on activated clay and observed that the rate of adsorption was highly affected by the degree of agitation when the agitation speed was increased from 200-600 rpm. On increasing the agitation rate beyond 400 rpm, the adsorption rate showed slight variations indicating that the film thickness had no significant effect beyond the agitation speed of 400 rpm.

Patel and Vashi[35] predicted that the continuous increment in percentage removal of dye was observed with increasing agitation speed up to 500 rpm and equilibrium was achieved at 500 rpm. Increasing the agitation speed beyond 500 rpm showed no effect on rate of adsorption. An increased adsorption rate at higher agitation speeds was probably due to increased mobility of adsorbate molecules. It is studied

the removal percentage of AR 97 (having concentration 50 mg L^{-1}) at different agitation speeds (80-180 rpm) within contact time of 1 h and observed that the rate of removal increased from 50.0-73.9% with increasing agitation speed. This can be explained by the fact that increasing the agitation speed increases the external film transfer coefficient by reducing the film boundary layer between dye molecules and surrounding particles[87].

Kisku *et al*.[99] also studied the effect of agitation speed (80-200 rpm) on percent dye removal of disperse blue 79:1 and disperse orange 25 onto CFA. The rate of removal for both dyes increased as agitation speed was increased up to 140 rpm. Disperse blue 79:1 and disperse orange 25 both showed maximum removal at 140 rpm. However, the net removal (i.e. percent change from 80-140 rpm) of DO at 25, 35 and 45°C was 44, 34 and 25% respectively, for DB, it was 39, 29 and 22% respectively and for disperse orange 25 it was 4.9, 3.9 and 2.9% higher than disperse blue 79:1.

Tiwari *et al*.[14] studied the removal of disperse blue 79:1 and disperse orange 25 onto modified cenospheres. Both the dyes showed maximum removal at 140 rpm. The net removal (i.e. percentage change, 80-140 rpm) of disperse orange 25 at 25, 35 and 45°C was 30, 23 and 20%, respectively, of disperse blue 79:1, was 24, 22 and 18%, respectively and the net removal of disperse orange 25 was 6, 1 and 2% higher than disperse blue 79:1.

Albroomi *et al*.[120] explained that the thickness of the liquid film surrounding a particle depends on the agitation speed and this film thickness also affects the mass transfer rate of a dye to a particle. They conducted a number of experiments at various degrees of agitation (25-125 rpm) for the adsorption of methylene blue and azo dye tartrazine on activated carbon. The results indicated that the rate of agitation influenced the removal efficiency when the agitation speed was increased from 25-125 rpm. At agitation rates higher than 100 rpm the removal efficiency showed very slight variation for the adsorption of methylene blue dye, indicating the negligible effect of film thickness beyond the agitation speed of 100 rpm, therefore, the agitation rate of 100 rpm was selected as the optimum value for all the future experiments.

Effect of contact time: Contact time is another important parameter which has been found to affect the sorption process significantly. The effect of contact time on the adsorption of congo red dye onto orange peel was studied by O'Mahony *et al*.[81]. It is revealed that the percentage adsorption increased initially for first 60 min and thereafter rate of removal slowed down. The equilibrium was seen at 90 min. Mall *et al*.[116] observed the effect of agitation time on

the removal of congo red and found that sorption rate was rapid in the first 15 min and optimum equilibrium contact was attained after 4 h. Akhtar *et al*.[103] reported the adsorption of 2,4-dichlorophenol on the surface of rice husk. They observed that percentage removal increased (26-97%) with increase in contact time up to 10 min. The equilibrium condition was achieved at contact times in between 10-20 min. Thereafter with further increase in contact time the removal 2,4-dichlorophenol was almost stagnant.

Ahmad *et al*.[121] also studied the effect of contact time onto palm ash to combat the problem of direct dyes and found that adsorption capacity increased with increase in contact time. The equilibrium condition was achieved after 1 h of contact time. The concentration of dye considered by the authors ranged from 50-600 mg L^{-1}. The uptake of dye increased from 32-321 mg g^{-1} with an increase in dye concentration. At first, rate of adsorption was high due to adsorption of dye molecules on the upper surface of palm ash. Then the rate decreased as the dye molecules penetrated into the inner porous structure of the palm ash. Wheat shells have also been used as an adsorbent for the uptake of direct blue 71 dye to find the effect of agitation time Bulut *et al*.[104]. The equilibrium condition was found after 36 h. Initially, it was observed that 50% adsorption occurred within 12 h i.e., rate of removal was rapid.

Ardejani *et al*.[117] studied almond shells as a potent low cost adsorbent for the adsorption of direct red 80 dye. The percent removal was initially fast and then became almost stagnant as the time passed. The maximum percent uptake of dye was 94% at equilibrium time 5 h. Mohan *et al*.[122] investigated *Spirogyra* sp., 102 for the adsorption of direct azo dyes. The uptake of dye was rapid in the beginning and then became almost constant at equilibrium point. Maximum dye was adsorbed in 2 h of the agitation time having dye concentration from 5-15 mg L^{-1} and percentage removal of dye was 64.0-35.3%. The removal of dye was rapid in the initial stages of the process due to association of binding sites on the biomass. Khaled *et al*.[123] used orange peel carbon as an adsorbent for the removal of direct yellow 12 varying the concentration from 25-125 mg L^{-1}. Initially, adsorption rate was high and about 75% adsorption took place in the first 19 min and then adsorption rate began to slow down. In the beginning, the high rate of adsorption might be ascribed to the presence of positive charge on the orange peel carbon which developed an interaction with negatively charged direct yellow 12 dye. Finally, the rate of adsorption began to slow down after 20 min due to slow movement of dye molecule into the interior of bulk of the orange peel carbon and equilibrium condition was reached after 2 h.

The adsorption of direct N-blue 106 dye was studied by El Nemr *et al.*[124] and it was observed that removal of dye increased with increase in the agitation time and about 2/3 dye was removed within 10 min and equilibrium condition was achieved after 3 h. Kurniawan *et al.*[85] investigated the removal of direct brown and direct brown 2 dyes varying the initial dye concentration from 330-900 and from 320-600 mg L^{-1}, respectively. The equilibrium adsorption capacity of direct brown dye remained constant after 2 h upto 3 h and for direct brown 2, the adsorption capacity remained constant after 3 h upto 5 h. The adsorption rate was found to increase quickly in the initial stages.

Ho *et al.*[125] showed that the adsorption capacity increased with increase in dye concentration and the rate of adsorption on the surface of adsorbent must be proportional to driving force times an area. A rapid initial removal of dye was reached within 30 min of contact time and adsorption capacity at equilibrium increased from 86.32-348.87 mg g^{-1}, while the percent removal decreased from 100-61% varying the methylene blue dye concentration from 200-1000 mg L^{-1}. With further increase in contact time (<30 min), the equilibrium condition was reached. The rapid removal of dye molecules was due to solute transfer, as there were only adsorbate-adsorbent interactions with almost no interference from solute-solute interactions by Kaczmarski and Bellot[126]. The initial rate of removal was therefore greater for high initial methylene blue dye concentrations, the resistance to the dye removal losing as the mass transfer driving force increased. Gupta *et al.*[89] studied the adsorption of orange DNA13 and vertigo blue 49 onto carbon slurry. The significant removal of dyes was observed in the first 20 min and equilibrium condition was reached in 60-90 min. However, for the sake of combined experiments, an equilibrium time of 90 min was measured to be optimum in further experiments.

Effect of particle size: The fact that there exists a direct relation between the surface area of adsorbent and its adsorption capacity makes particle size of adsorbent a very important parameter. The relation between particle size and the percent removal of direct dyes was studied by Gong *et al.*[112]. They examined that adsorption capacity of the adsorbent increased with decrease in particle size. The effect of different sizes of the adsorbent particles on the removal of Congo red dye was analyzed by Jain and Sikarwar[118]. They found that adsorption increased with the decrease in adsorbent particle size for both activated carbon and sawdust biomass. The maximum adsorption (94.5% for activated

carbon and 76.6% for sawdust) was observed at the nominal particle size (<106 BSS mesh). This may be attributed to the increased surface area offered by the smallest particle size of adsorbent.

Kaczmarski and Bellot[126] conducted adsorption experiments using various particle sizes (40-125 μm) and varying the adsorbent dosages (500-15000 mg L^{-1}) of the fly ash at pH 6.0, initial concentration of 100 mg L^{-1}, temperature of 22°C and agitation speed of 250 rpm. The percentage removal at equilibrium decreased with increase in particle size and increased with increasing adsorbent dosage. With smaller adsorbent particle the rate of adsorption is relatively higher and this is because smaller particles offer larger surface areas and more adsorption sites or in other words surface to mass ratio increases.

Sutar and Ranade[127] also found an inverse relation between particle size of adsorbent and rate of adsorption. Maximum percentage removal (100%) was observed for the particle size of 0-0.25 mm. The removal percentage of 95.49, 94.81 and 81.60 were obtained for particle size varying from 0.25-0.50, 0.50-0.75 and 0.75-1.00 mm, respectively. As size 0-0.25 mm gave best result among all the considered sizes, this sized adsorbent were used for other experiments.

Disperse blue 79:1 (DB) and disperse orange 25 (DO) dyes, having an azo group are toxic when they enter the environment in a quantity or concentration that has direct or enduring harmful effect on the environment as well as aquatic ecosystem. These dyes are also used for sublimation printing of synthetic fibers and are the colorant used in crayons, inks and commercially sold as "Iron-on transfers". Mostly, mixture of different dyes is used for final color of textile[14]. Many dyes often contain not only the main component, but also several impurities[128]. Colored substances present in aquatic living organisms have been associated with changes in protozoan colonization rate, phytoplankton species composition and primary productivity. In addition, secondary production, macro-invertebrate behavior and macro-invertebrate community system can also be affected. Color can also alter the availability and toxicity of heavy metals to fishes[100]. Ions of disperse dyes in the water streams either reflect back the solar radiation or scatter in the water bodies. Solar radiation cannot penetrate beyond littoral zone and therefore, the deep water zone (i.e., aphotic zone) develops anaerobic conditions. The entire ecological cycle including self-purification system of water stream is disturbed due to lack of dissolved oxygen. Dye is also associated with other organic toxins such as wood extracts and chlorinated organic substances formed during the bleaching stages.

The specific surface area can be calculated using BET method having saturated pressure 760 mmHg. High surface area of adsorbents has the structures of small particle sizes, with medium surface areas and whose mesopores are the principal contribution to their porosity. Overall, for more adsorption occurs on the surface of adsorbent, the surface to mass ratio should be more, which provided more site of adsorption of pollutants. A good adsorbent should generally possess a porous structure (resulting in high surface area) and the time taken for adsorption equilibrium to be established should be as small as possible so that it can be used to remove dye wastes in lesser time.

CONCLUSION

Presently wastewater treatment is emerging field for research due to increasing rate of population with respect to limited resources. Water pollution is heavily generated by anthropogenic activity including industrial discharge which are unable to treat the colored wastewater coming out from various process outlets due to high cost of treatment. Textile mill's colored effluent have some desired properties which resist lighting, oxygen, acids and bases present in wastewater which cause harm to aquatic organisms or humans by mutagenic and carcinogenic effects. Wastewater treatment and reuse is necessarily in regards to water balances and management. In this study, we attempted to summarized the use various adsorbents, abundantly available and modified precursors for adsorption of disperse dyes from aqueous solution. It is great challenges to treat textile wastewater, especially when the wastewater have more than one component and study of the possible interactions between different chromophores is very useful for the treatment of real effluents. During the investigation, decolorizing efficiencies based on various essential factors like pH, agitation speed, adsorbent dose, adsorbate concentration, temperature, contact time were noted. Due to the anticipated water scarcity, the concern must be taken up on priority for wastewater treatment. An effort has been made to review the efficiency of various adsorption techniques used for dye removal from colored wastewater and the use of economical techniques for successful wastewater treatment and management.

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