Chapter 16
Nanotechnology: A New Scientific Outlook for Bioremediation of Dye Effluents

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Contents

16.1 Introduction 355
16.2 Dyes and Their Classification 357
16.3 Toxicology Effects of Dye 357
  16.3.1 Acute Toxicity of Textile Dye 358
  16.3.2 Chronic Toxicity of Textile Dye 358
16.4 Nanotechnology for Textile Dye Effluent Remediation 360
  16.4.1 Nano-adsorbents 360
  16.4.2 Nano-catalysts 363
  16.4.3 Nanofiltration Membranes 364
16.5 Mechanism for Textile Dye Effluents Adsorption 364
16.6 Advantages and Disadvantages 365
16.7 Conclusion 366
References 366

16.1 Introduction

Industrial wastewater generation and its treatment are a worldwide problem. Effluent is treated or untreated wastewater that flows out of a treatment plant, sewers, or industrial outfall. Although industries manufacture various useful products for mankind, they also generate different waste by-products in various forms that are responsible for the exaction of hazards and pollution. Most of the waste products are discharged in the soil and water bodies that ultimately pose a serious warning to mankind and routine functioning of the ecosystem (Chaves et al. 2016). Currently, the existence of hazardous pollutants in industrial waste effluents is the most serious environmental problem. The untreated wastewater discharge is greatly affecting marine ecosystem, livelihoods, and food chain. According to 2017 world water development report, it is estimated that worldwide 70–80% of industrial effluents

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R. Prasad, E. Aranda (eds.), *Approaches in Bioremediation*, Nanotechnology in the Life Sciences, https://doi.org/10.1007/978-3-030-02369-0_16

are disposed into the environmental surroundings without any prior treatment. Main contributors of toxic industrial effluents are the by-products coming from the various industries such as textile, paper, dying, chemicals, fertilizers, pesticides, food processing, mining, etc. Dyes are organic colorants that contain unsaturated compound (chromophores) and functional group (auxochromes). The chromophores are responsible for color formation, and auxochromes intensify the color of dye. There are various chromophoric groups such as carbon monoxide (–CO), reactive oxygen (O), nitrogen (N-), nitrogen oxide (-NO_2), and quinonoid ($C_9H_{13}N_5O_3$), while auxochromes include aldehydes (–CHO), carboxylic group (–COOH), hydroxyl group (-OH), sulfonic acid (-SO_3H), and –NHX_2 (Solis et al. 2012). Dyes are divided into different categories based on chromophoric groups, viz., "azo (-N=N-), acidic, basic, disperse, reactive, and anthraquinone dyes." Natural dyes have constricted spectrum of colors and tendency to fade quickly when brought to sun exposure, and cleansing has restricted their application (Firmino et al. 2010). Conversely synthetic dyes provide an immense spectrum of colors that are bright and color fast. Among the synthetic dyes, azo dye is the most commonly used group. The disposal of effluents originating from various industries into water pools and circumambient is of major concern (Elango et al. 2017). Among these, the textile industry is considered as the largest contributor to dye wastewater, because it is censurable for two thirds of overall production of dye wastewater. During dyeing process, a large proportion approximately 50% of the initial dye load remains unfixed or unconstrained and released in environment as effluents (Singh and Arora 2011). It is estimated that 5000 tons of dyeing effluents are disposed directly or indirectly to the environment without any adequate treatment every year. Even at a minute concentration, colorants in wastewater are unpleasant and very threatening to aquatic flora, fauna, and human lives (Mahmoodi and Arami 2009). In India, dye production is estimated to be around 60,000 tones/year or 6.6% of the world production (CPCB, India). Generally, textile wastewater contains dye concentration in the vicinity of 1 g L^{-1} (Tan et al. 2000). The color-free effluents' discharge to water bodies has made treatment of textile industrial wastewater a top priority. Therefore, it is of topmost importance to treat dye-containing effluents.

Conventionally, there are various chemical, physical, and biological methods for treatment of colorants from wastewater effluents including surface adsorption, membrane filtration membranes, coagulation, ozonation, photocatalyst-based oxidation, and Fenton process (Singh et al. 2012). Among these methods, adsorption process is most efficient and globally used as it is low cost, flexible, easy and produce less harmful by-products (Qu et al. 2013). Recent advances in nanotechnology offer incredible potential for the remediation of wastewater. Nano-bioremediation process involves the reduction of pollutants from contaminants by using nanoparticles, which is synthesized by green waste with the aid of nanotechnology. The green method has evolved remarkably to synthesize the novel nanomaterials that are stable, cheap, and eco-friendly. There are various conventional methods that are available to manufacture the nanomaterials, but the green route for synthesizing is more effective and advantageous. The ease of synthesis, less toxic nature, and environment-friendly approach of biological method make it more adoptable process

(Mie et al. 2014). Nowadays, nanomaterials have been proved as more efficient and more exploit for wastewater treatment because of their large surface area and economical synthesis. The oxide-based nanomaterials have been extensively explored for remediation of dye-containing effluents. The iron oxide nanoparticles are more favorable than other nanoparticles due to having super paramagnetism property. Super paramagnetic nanoparticles have many advantageous features such as biocompatibility, high surface area to volume ratio, less toxicity, chemically inert, and small diffusion resistance. The surface behavior of nanoparticles can be modified with some functional groups and organic or inorganic ions, which impart surfaces having good potential for removing or decolorizing the dye effluents (Huang et al. 2012). Thus, nanotechnology offers highly efficient and multifunctional processes that render high-geared and economical wastewater treatment techniques that are less dependent on large infrastructure (Qu et al. 2013).

16.2 Dyes and Their Classification

A dye is an organic colored substance that has strong or specific affinity with substrate on which it is being poured and may alter any crystal formation of the colored organic materials temporarily. The colorants adhere to their compatible substrate by forming complexes via covalent bond. Dyes are divided into various types according to their solubility, chemical structure, and application (Fig. 16.1). It is measured that over 10,000 different colorants are used in textile industries and around 7×10^5 tons of synthetic colorants are produced yearly.

16.3 Toxicology Effects of Dye

Textile dyes are important and globally being encountered directly or indirectly in many aspects of daily life. The first synthetic dye mauveine was invented by William Henry Perkin in 1856. The natural sources of colorants are plants, insects and molluscs, but narrow range of colors and tendency to fade quickly when exposed to the sun and cleansing have restricted their application (Shah et al. 2013). Conversely, synthetic colorants provide a broad spectrum of colors that are bright and color fast. After many years, various synthetic dyes were manufactured by workers such as fuchsine, auramine, benzidine, and naphthylamine and found that these are carcinogens for humans. It has been estimated that annually a million tons of synthetic dyes are produced at the international level. Textile wastewaters containing mixture of dyes generally have high biological oxygen demand (BOD), chemical oxygen demand (COD), total soluble solids (TSS), total dissolved solids (TDS), and alarming toxicities (Casieri et al. 2008). The textile industrial revolution aroused a great threat of pollution. The toxic effect of textile dyes can be classified into two types of effects, i.e., short-term (acute) effect and long-term (chronic) effects.

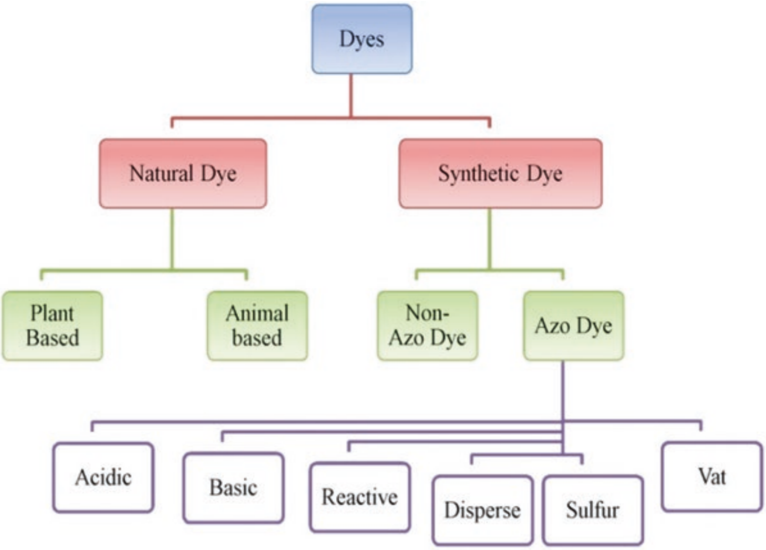

Fig. 16.1 Classification of dyes

16.3.1 Acute Toxicity of Textile Dye

Acute toxic effect involves skin irritation, skin sensitization, eyes irritation, oral ingestion, and inhalation. Acute toxic effects are mainly caused by reactive dyes and disperse dye (Tronnier 2002). The hydrophilic reactive dyes contain a group that forms a complex or covalent bond with the hydroxyl groups (-OH) present in the fibers during the dyeing activities. Reactive dyes are problematic to the workers who manufacture or handle the dyes. There are many evidences of toxicity caused by reactive dye such as contact dermatitis, allergic conjunctivitis, asthma, and other allergic reactions. The reactive dyes have capability to react with HSA (human serum albumin) and form a dye-HSA conjugate that acts as an antigen. The specific immunoglobulin E (IgE) produced by that antigen which forms histamine compounds that leads to human allergic reactions (Hunger and Sewekow 2003).

16.3.2 Chronic Toxicity of Textile Dye

Genotoxic textile chemicals include dye that are mutagens, carcinogens, and teratogens and pose long-term health hazard (Table 16.1). Mutagen chemicals create mutations, but they may or may not be carcinogens to humans and animals. Teratogens are rarely used in textile industries because it causes birth defects in the offspring although teratogenicity is very uncommon in textile dyes. Some dyes such as 4-aminoazo dyes were found to be carcinogen to animals, and 2-naphthylamine (water soluble) cationic dye is a potent human bladder cancer-causing agent

(Myslak and Bolt 1988). Some dyes have capability to leach out an aromatic amine, a rodent carcinogen. Around 500 azo dyes form the aromatic amines after reduction of azo functional groups. Anthraquinone dye is the most important class of dyes that contains amino or methyl-amino groups that are mutagenic and carcinogenic, for example, 4-aminobiphenyl, benzidine, and toluidine, which are proven to be human carcinogens. Some disperse dyes such as disperse orange and disperse blue are

Table 16.1 List of toxic dyestuffs

S.No.	Compound name	CAS No.	Toxic properties	Usage
1	Disperse dye			
	Disperse Blue 1	2475-45-8	Carcinogenic	Dyeing of polyester
	Disperse Orange 11	82-28-0		
	Disperse Yellow 3	2832-40-8	Allergic	
	Disperse Yellow 23	6250-23-3		
	Disperse Orange 149	85136-74-9		
	Disperse Red 60	17418-58-5	Mutagenic	
2	Acid Green 16	12768-78-4	Mutagenic	Dyeing of cotton
3	Acid Red 26	3761-53-3	Carcinogenic	Dyeing
4	Basic Green 4	10309-95-2	Mutagenic	Dyeing
5	Basic Red 9	569-61-9	Carcinogenic	Dyeing
6	Basic Violet 14	632-99-5	Carcinogenic	Dyeing
7	Direct Black 38	1937-37-7	Carcinogenic	Dyeing
8	Direct Blue 6	2602-46-2	Carcinogenic	Dyeing
9	Direct Red 28	573-58-0	Carcinogenic	Dyeing
10	Amines		Carcinogenic	Dyeing of cotton, silk, wool
	Benzidine		Bladder cancer	
	2-Naphthylamine			
	4-Aminodiphenyl			
	Aminoazotoluene			
11	Formaldehyde		Suspected carcinogen, skin and respiratory sensitizer	Dye fixing
12	Alkylphenol ethoxylates		Aquatic toxicity, endocrine disruption	Wetting, washing
13	Chlorophenol		Cancer, skin sensitization	Dry cleaning
14	Heavy metals (Cd, Pb, Ni)		Nephrotoxic	Dyes
15	Volatile organic compound		Eye damage and respiratory sensitization	Printing

carcinogen, while disperse violet is mutagenic to some extent (Hunger and Sewekow 2003). Aromatic amines are the precursors for many textile colorants and responsible for the carcinogenicity. Almost all nitrosamines are carcinogens. Hydrazines are applied in textile industries to produce heterocyclic coupling components having carcinogenic properties.

16.4 Nanotechnology for Textile Dye Effluent Remediation

Nanotechnology is the branch of science that addressed the dimensions and tolerance of less than 100 nm, especially the manipulation of matter on the basis of atoms, molecules, and supramolecules scale. Thus, nanotechnology is characterized by the use of very ultrafine manufactured particles (<100 nm) called ultrafine particles or nanomaterials. Nanomaterials are atoms or molecule aggregates with size range between 1 and 100 nm that can drastically modify their physiochemical properties compared with the bulk material. The more reactive forms of nanomaterials are membranes, nanowires, tubes, films, particles, quantum dots, and colloids. The nanomaterials are broadly classified into two groups of organic and inorganic nanoparticles. Organic nanomaterials include carbon nanoparticles, and inorganic nanomaterials include magnetic nanoparticles, noble metal nanoparticles (e.g., silver and gold), and semiconductor nanoparticles (e.g., titanium oxide and zinc oxide). The removal of pollutants from environment with the help of nanomaterials synthesized by using biological waste is called nano-bioremediation (Yadav et al. 2017).

In general, nano-bioremediation involves the use of nanomaterials either in in situ (within) or ex situ (off place) treatment of contaminated material (Latha and Gowri 2014). Due to the some negative sides of other traditional methods, green method is widely used for synthesis of nanoparticles. Biological route for synthesis of nanoparticles is eco-friendly, cost-effective, and stable and has grown markedly to create novel materials (Prasad et al. 2018). Microbial nanotechnology is also a newly emerged route to produce a nanoparticulate catalyst that involves the precipitation of transition metals such as palladium, gold, and iron on bacterial cell wall, resulting in the formation of bionano-material (Johnson et al. 2013). The efficiency of degradation of contaminants can be enhanced by combining different treatment technologies. Recently nano-bioremediation has been suggested as efficient, eco-friendly, and economically cheap alternative for both resource conservation and environmental remediation. The nanotechnology for wastewater treatment can be classified into three main categories on the basis of nature of nanomaterials: nano-adsorbents, catalysts, and membranes.

16.4.1 Nano-adsorbents

In recent years, nanoparticles are being used as potent adsorbents. The nanoparticles due to having small size have high surface area to volume ratio which increases the adsorption capacity and chemical activity of nanomaterials for adsorption of metals on their surface (Kalfa et al. 2009). The two major factors that affect the adsorption phenomenon are adsorption coefficient K_d and recitation partitioning of pollutants (Hu et al. 2010). The nanoparticles such as zinc oxides, magnesium oxide, titanium oxide, ferric oxide, and carbon-based nanomaterials have been used for contaminants adsorption (Tyagi et al. 2017). Nano-adsorbents possess two main properties that are innate surface and external functionalization. The large surface area to volume ratio, adsorption capacity, pH, temperature, chemical reactivity, location of atoms, incubation time, and high binding energy on surface are important factors that affect the adsorption rate (Khajeh et al. 2013). The nanoparticles used in remediation process of wastewater must be nontoxic and high adsorption and desorption capacity.

16.4.1.1 Oxide-Based Nano-adsorbents

Oxide-based nano-adsorbents are organic and inorganic ultrafine materials which are usually synthesized by utilizing metals and nonmetals. These include titanium oxides, iron oxides, zinc oxides, magnesium oxides, and manganese oxides (Tyagi et al. 2017). The natural occurrence of metal and nonmetal oxides and its simple synthesis process make them cheap materials for the removal of wastewater contaminants. The iron oxide nanoparticles such as α-Fe_2O_3 are found to remove most of the orange II (a common azo dye) at room temperature. The decolorization of dye is due to the electrostatic force of attraction between the iron oxide surface and orange II (Fig. 16.2). Furthermore, the "catalytic combustion at 300 °C in air for 3 h regenerate the iron oxide containing Orange II dye and the regenerated a-Fe_2O_3 material have almost the same adsorption rate" (Zhong et al. 2006). The Fe_3O_4 nanoparticles incorporated with polylysine (Fe_3O_4@GPTMS@P-Lys) absorb methylene blue, and Acid Red 18 was studied by Zhang et al. The copper oxide nanoparticles in association with activated carbon (Cu-NP-AC) under pH 2, contact time 25–30 min, adsorbent dosage (0.01–0.06 g), and temperature 333 K were found as potential adsorbent for removal of Acid Blue 129 (Nekouei et al. 2015). The ability of $MnO_5CuO_5Fe_2O_4$ nanospinels to adsorb brilliant green from textile effluents was analyzed by Hashemian et al. (2015). Amorphous metal oxide nanoparticle (Fe_2O_3, CoO, NiO) fabrication via a green process (laser irradiation in liquid) and their ability to remove methylene blue was tested by Li et al. (2015).

16.4.1.2 Carbon-Based Adsorbents

Activated Carbon

Carbon-based nanomaterials have been proved as more advantageous than available materials due to having manageable pore size and large surface area. Activated carbon is the most frequently used method for dye decolorization by adsorption. Carbon materials are very effective for adsorbing cationic, reactive, disperse, and

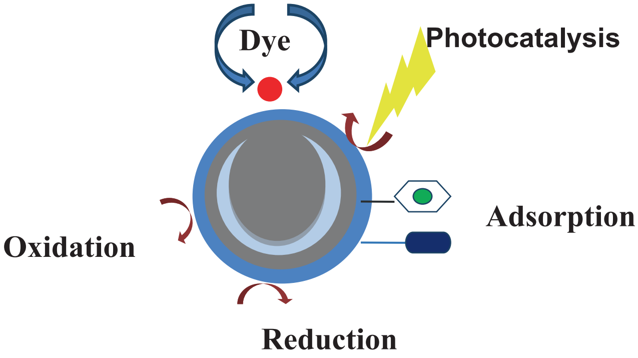

Fig. 16.2 Different surface activities of oxide nanoparticles

acid dyes (Rao et al. 1994). Nano-MgO-encapsulated activated carbon was used for the removal of carcinogenic (CR) dye (Daniel and Syed 2015). Adsorption efficiency depends upon the type of carbon used and the characteristics of the effluents. Dye removal efficiency can be improved by using doses of carbon. This adsorption phenomenon has some drawbacks such as carbon that is expensive and has to be reactivated after treatment. Reactivation of activated carbon results in 10–15% loss of the sorbent.

Carbon Nanotube

Carbon nanotubes are emerging potent adsorptive materials that have a great potential to remove contaminants from wastewater through adsorption because of its high surface area, cylindrical hollow structure, hydrophobic walls, and flexible surfaces (Lu et al. 2006). From the researches, it has been analyzed that single-walled carbon nanotubes (SWCNTs) show higher adsorption capacity than multi-walled carbon nanotubes (MWCNTs). The hexagonal arrangements of carbon atoms allow them to interact with other molecules or atoms through hydrophobic and π-π electronic interaction. The batch experiment studies to analyze adsorption rate of cationic methylene blue (MB) and anionic orange II (OII), from aqueous solution by using multi-walled nanotubes (MWNTs) and carbon nanofibers (CNF) as adsorbents, were analyzed by Rodríguez and coworkers (Rodriguez et al. 2010). Yao et al. reported 41.63 mg/g adsorption capacity of carbon nanotubes (CNTs) for removal of methylene blue at 333 K. Some researchers performed a comparative study of multi-walled carbon nanotubes (MWCNTs) and single-walled carbon nanotubes (SWCNTs) as nano-adsorbents for the removal of reactive blue 4 (RB4) textile dye from aqueous solutions as a function of effects of pH, shaking time, and temperature (Machado et al. 2012). Magnetic-modified multi-walled carbon nanotubes (MWCNTs) were found effective for removal of cationic dyes crystal violet (CV), thionine (Th), Janus green B (JG), and methylene blue (MB). Impregnation of chitosan beads with carbon nanotubes was investigated as more efficient adsorbent for dye removal (Chatterjee et al. 2010).

16.4.2 Nano-catalysts

The inorganic nano-catalysts such as semiconductor and metal oxides are gaining a noticeable attention for water and wastewater treatment. Different kinds of nano-catalysts are used for remediation, for example, photocatalysts, electrocatalysts, and Fenton-based nano-catalysts for improving chemical oxidation of pollutants (Table 16.2). Photocatalytic oxidation is the latest remediation technique. It utilizes ultraviolet (UV) illumination and titanium oxide (TiO_2) and generates low waste. The UV light helps to enhance the photocatalytic activity of TiO_2 during dye degradation. Among various nano-catalysts, TiO_2 is highly applied in photocatalytic reaction due to its high reactivity and chemical balancing (Akhavan 2009). Zinc oxide has been found to be well suited for photocatalysis of dye effluents, as it contains broad bandgap like TiO_2 (Lin et al. 2014). The use of nano-catalysts in chemical oxidation process has some advantages such as short period of time, target recalcitrant substances, and efficient transformation ability. The nano-catalysts are beneficial, but also have some negative aspects such as high cost of nanoparticles and inadequate reusability.

Table 16.2 Photocatalytic efficiency of nano-catalysts for removal of dye from textile effluents

Catalysts	Light region (nm)	Pollutant	Results	References
Ti_2CO coating	420	Methylene blue (MB)	Removal of dye from 10 to 8.5 μ mol/L	Guan et al. (2016)
TiO_2/trititanate	UV-vis region	Rhodamine B (RB)	The degradation efficiency >91% was achieved	Chen et al. (2015)
AgBr/Zno	410	Methylene blue (MB)	Up to 87% MB was decomposed after 240 min	Dai et al. (2014)
3D SnO	365	Methyl orange (MO)	Photocatalytic degradation of MO was 83% after 150 min	Cui et al. (2015)
ZnO nanorods	365	Rhodamine B (RB)	The quenched catalyzed improved the photocatalytic degradation of RB	Fang et al. (2015)
Zero-valent nanocopper	420	Methyl orange	35% degradation of dye was achieved within 80 min	Liu et al. (2016)
Biomorphic TiO_2 photonic crystal	>420	Methyl orange	Up to 30% removal of dye was observed within 4 h	Wang et al. (2016)
$CuFe_2O_4@C_3N_4$	>420	Orange II	Around 98% of orange II is removed within 210 min	Yao et al. (2015)
Polyaniline ZnO	Visible region	Methylene blue and malachite green	99% dye degradation was achieved under sunlight	Eskizeybcket al. (2012)
$Y_2O_3Eu^{3+}$	350–400	Methylene blue	Up to 90% degradation efficiency of MB was observed	Ramgopal et al. (2015)
ZnO/Zn	365	Methylene blue	98% dye degradation was achieved	Lin et al. (2014)

16.4.3 Nanofiltration Membranes

Membrane filtration technology is the current advanced wastewater treatment technology. Nano-membranes are fabricated by nanoparticles to enhance the remediation efficiency in the aspect of permeability, catalytic reactivity, and fouling resistance (Zhang et al. 2015). Membrane filtration technology is simple, economical, effective remediation, and low space requirement. Nano-membrane filtration technology is highly used for removal of dye and other contaminants from wastewater effluents (Jie et al. 2015). The literature review clearly indicates the great potential of nanofiltration membrane for textile effluents treatment. Some researchers reported the NF membranes exhibit good performance for dye removal. Lopes et al. observed the maximum dye rejection up to 95–99% by using nanofiltration membrane NF45 and DK1073 (Lopes et al. 2005). The performance of nanofiltration membrane MPS 31 was investigated for dye retention that varied from 90% to 97%. Sungpet and coworkers investigated the higher efficiency of secondary layer formed by MPF 36 due to having larger MWCO in the presence of reactive dye and chloride (Sungpet et al. 2004). Tang and Chen hypothesized the dye rejection is independent on dye concentration. They found that at a pressure of 5 bar if the concentration of dye gradually increases from 92 to 1583 ppm, the dye rejection rate remains constant (Tang and Cheng 2002). The efficiency of membrane for decolorization of effluents is also dependent on cross-flow velocity. Although the nanofiltration is advantageous, membrane fouling is a weakness of filtration technique. Dyes introduce the undesirable flux decline by forming a colloidal fouling layer. The membrane fouling can be reversed by using feed pre-treatment modification of membrane surface and by controlling membrane cleaning.

16.5 Mechanism for Textile Dye Effluents Adsorption

Adsorption phenomenon of dye depends upon three main parameters, i.e., solution osmolarity, nature of adsorbents and adsorbates. Thus, the factors which affect the adsorption phenomenon are size of the adsorbate and adsorbent, charge on the surface of adsorbate and adsorbent, and pH and temperature of the solution. Generally, the redox chemical reactions take place on adsorbent surface, and the force responsible for adsorption includes electrostatic force of attraction, π-π bonding, hydrophobic interaction, and covalent bonding. There are various functional groups such as –OH, -COOH, and C-O which are present on adsorbent surface that provide active sites for binding of dye molecules on the surface (Shim et al. 2001). Figure 16.3 shows the ion exchange reaction as membrane coated with nanoparticles comes in contact with dye molecule. The proton (H^+) from water molecule after the dissociation of hydrogen bonding was exchanged with the dye molecule that finally leads the adsorption of dye on nanoparticles surface.

16.6 Advantages and Disadvantages

Nanoscience is an emerging technology that has great potential for effective treatment of wastewater. The high adsorption capacity, less waste by-product generation, and low toxicity of oxide-based nanoparticles and nanofiltration membrane make them more affirmative for removal of dye-containing textile effluents. The synthesis and utilization of nanoparticles have been widely used due to their small size (nano range), easy synthesis, modification ability, biocompatibility, and high surface area to volume ratio (Prasad et al. 2016, 2018). The green route for synthesis of nanoparticles makes them more accessible, eco-friendly, and low-cost adsorbents for dye removal Aziz et al. 2015. Along with the advantageous properties, the nanoscience has some drawbacks such as less stability, high reactivity with any functional group, antifouling of membranes, and leaching of by-products in environment that directly affect the flora and fauna. The advance researches are required to enhance the stability of nanoparticles by reducing their surface energy, high rejection rate, and antifouling resistant for nano-membranes. Thus, it is utmost important to make nanotechnology more clean, cheap, and environment-friendly.

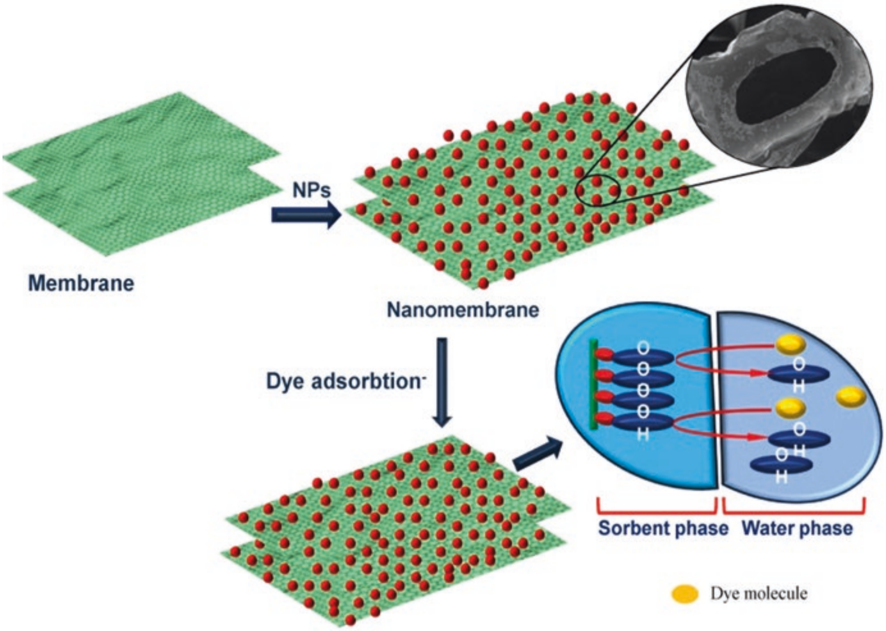

Fig. 16.3 Mechanism for adsorption of textile dye effluents on nanoparticles

16.7 Conclusion

In present scenario, there is a great need of advanced technology for wastewater treatment to ensure better quality of water and eliminate chemical and biological contaminants. Thus, nanoscience is an emerging promising technology for advanced wastewater effluent treatment. This chapter illustrates the various nanomaterials that have been developed and investigated successfully for advanced treatment of wastewater effluents. The chapter focused the nanomaterials such as nano-adsorbents, nano-catalysts, and nanofiltration membrane for degradation of dye-containing wastewater effluents. The nano-adsorbents and nano-catalysts were found to have more specific pollutant removal efficiency. Indeed, nanomaterials are more efficient than any conventional method for advanced treatment of wastewater, but the technology has some negative side too that has to be resolved. Thus, more researches are required to re-evaluate the toxicity efficiency of advanced nanomaterials in order to make it more clean and eco-friendly.

Acknowledgments We acknowledge the ministry of human resource and development (MHRD), India, for providing financial assistance under the research project "Center for Excellence (CoE) – Water and Energy." Authors are deeply grateful to Prof. Aditya Shastri for giving the research amenities and the Bioinformatics Centre, Banasthali Vidyapeeth, Rajasthan (India) for use of computational facilities.

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