GREEN CHEMISTRY FOR DYES REMOVAL FROM WASTE WATER

Research Trends and Applications

Edited by

Sanjay K. Sharma

Scrivener Publishing

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Green Chemistry for Dyes Removal from Wastewater

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Co-published by John Wiley & Sons, Inc. Hoboken, New Jersey, and Scrivener Publishing LLC, Salem, Massachusetts.
Published simultaneously in Canada.

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Cover design by Russell Richardson

Library of Congress Cataloging-in-Publication Data:

ISBN 978-1-118-72099-8

Printed in the United States of America

10 9 8 7 6 5 4 3 2 1

This book is for Kunal – Kritika….my twin angels on their 15th Birthday, with love.

Contents

Preface

Writing a preface for a book always has been a challenge as things are to be looked upon not only from the eyes of an editor, but also from a reader's perception and expectations; all the while keeping in mind not to do any injustice to the zeal of a contributor who has worked so hard to pen the text.

"Green Chemistry" two decade's old philosophy, has been attracting the attention of scientists worldwide. Academicians as well as industrialists are equally interested in this *new* stream of chemical science. Researchers, all over the world, are conducting active research in different fields of engineering, science and technology by adopting green chemistry principles and methodologies to devise new processes with a view towards helping, protecting, and ultimately saving the environment of our planet from further anthropogenic interruptions and damage. Achieving sustainability and renewability of resources is the basic spirit of green chemistry; it inspires us to try alternative "green" approaches in place of traditional "gray" practices in everyday industrial and scientific activities.

Water pollution is a matter of great concern. It's quality and potability is equally important for both domestic purposes and industrial needs. But, at the same time, industrial effluents pollute the available water resources. Dyes, as one of the pollutants, cause various serious health hazards and socioeconomic problems. It spoils the "productivity" of soil; which in turn may be the reason for other related issues, especially in developing countries. Removal of dyes from water or wastewater is therefore an important task. But, removing dyes at a cost to the environment should be avoided when considering which technique to use. So, the far important challenge is to make a removal technique sufficiently "green."

Water pollution is often discussed with respect to various pollutants and their treatments, but water pollution due to the presence of synthetic dyes has not been discussed sufficiently in the literature. So, the treatment of wastewater produced from industries using dyes (directly or indirectly) has tremendous scope worldwide. That is why dye removal is an important issue which needs to be addressed seriously.

The chapters in this book are the outcome of the scholarly writing of researchers of international repute with stellar credentials, who have tried to present an overview of the problem and its solution from different angles. These problems and solutions are presented in a genuinely holistic way using valuable research-based text from world-renowned researchers. Discussed herein are various promising techniques to remove dyes, including the use of nanotechnology, ultrasound, microwave, catalysts, biosorption, enzymatic treatments, advanced oxidation processes, etc., all of which are "green." The book contains eleven chapters, all of which focus on the theme of green chemistry and discuss tools and techniques which are eco-friendly, non-hazardous and, moreover, low waste generating.

The textile industry produces a large amount of dye effluents which are highly toxic as they contain a large number of metal complex dyes. The use of synthetic chemical dyes in various industrial processes, including paper and pulp manufacturing, plastics, dyeing of cloth, leather treatment and printing, has increased considerably over the last few years, resulting in the release of dye-containing industrial effluents into the soil and aquatic ecosystems. The textile industry generates highly polluting wastewaters and their treatment is a very serious problem due to high total dissolved solids (TDS), presence of toxic heavy metals, and the non-biodegradable nature of the dyestuffs present in the effluent. There are many processes available for the removal of dyes by conventional treatment technologies including biological and chemical oxidation, coagulation and adsorption, but they cannot be effectively used individually. Different types of dyes, their working and methodologies and various physical, chemical and biological treatment methods employed so far are comprehensively discussed in Chapter 1.

Adsorption is widely acknowledged as the most promising and efficient method because of its low capital investment, simplicity of design, ease of operation, insensitivity to toxic substances and ability to remove pollutants even from diluted solutions. In recent years, nanotechnology has introduced a myriad of novel nanomaterials that can have promising outcomes in environmental cleanup and remediation. Particularly, carbon-based nanomaterials such as carbon nanotubes and graphene are being intensively studied as new types of adsorbents for removal of toxic pollutants from aquatic systems. This extraordinary interest stems from their unique morphology, nanosized scale and novel physicochemical properties. Thus, Chapter 2 focuses on the use of nanotechnology in the treatment of dye removal.

Textile dyeing industries expend large volumes of water, which is ultimately discharged with intense color, chemical oxygen demand (COD), suspended/dissolved solids and recalcitrant material as unfixed dye residuals and spent auxiliaries. A typical reactive dyebath effluent contains 20–30% of the input dye mass (1500–2200 mgL^{-1}) and traces of heavy metals (i.e., cobalt, chromium and copper) that arise from the use of metal-complex azo dyes. The challenge to destroy dye residuals in biotreated wastewater effluents seems to be resolved by the introduction of advanced oxidation processes (AOP), whereby highly reactive hydroxyl radicals are generated chemically, photochemically and/or by radiolytic/sonolytic means. Hence, AOPs not only offer complete decolorization of aqueous solutions without the production of huge volumes of sludge, but also promise a considerable degree of mineralization and detoxification of the dyes and their oxidation/hydrolysis byproducts. The potential of ultrasound as an AOP is based on cavitation phenomenon, i.e., the formation, growth and implosive collapse of acoustic cavity bubbles in water and the generation of local hot spots with very extreme temperatures and pressures. Application of AOPs in dye removal is comprehensively discussed in Chapters 3 and 7.

The heterogeneous photocatalysis process has shown huge potential for water and wastewater treatment over the last few decades. Chapter 4 summarizes the photocatalytic oxidation process for dye degradation under both UV and visible light, application of solar light and solar photoreactor in dye degradation, and then finally discusses the dependence of different parameters (pH, photocatalyst loading, initial dye concentration, electron scavenger, light intensity) on dye degradation.

Several technologies have been developed to treat dye-containing effluents (DCEFs) such as coagulation-flocculation, filtration, sedimentation, precipitation-flocculation, electrocoagulation-electroflotation, biodegradation, photocatalysis, oxidation, electrochemical treatment, membrane separation, ion-exchange, incineration, irradiation, advanced oxidation, bacterial decolorization, electrokinetic coagulation and adsorption on activated carbon. From an industrial viewpoint, no single process provides adequate treatment, being that significant reduction of expenses and enhancement of dye removal can be achieved by the combination of different methods in hybrid treatments. "Biosorption" can be employed to treat DCEFs because it combines the advantages of adsorption with the use of natural, low-cost, eco-friendly and renewable biosorbents. Biosorption of organic dyes and related research opportunities and challenges are beautifully discussed in length in Chapters 5 and 8.

The enzymatic process using ligninolytic enzymes, such as laccases and peroxidases, is a relatively new emerging technology for the degradation of xenobiotics, including synthetic dyes in textile wastewater. This unique process employs a hybrid of chemical and biological oxidation using a combination of crude or purified enzymes from plant materials or fungal cultures as a biocatalyst and dissolved molecular oxygen or hydrogen peroxide as a chemical oxidant. This enzymatic process has a number of advantages over conventional physical, chemical and biological processes. Chapter 6 provides a comprehensive literature review on the enzymatic treatment of various synthetic dyes and discusses the recent progress and challenges associated with this technology. In addition, the fungal treatment of synthetic dyes and contaminated effluents, as well as the enzymology of the key ligninolytic enzymes, are covered in this chapter to explore the important roles of fungal enzymes in synthetic dye decolorization.

Adsorption is one of the best treatment methods due to its flexibility, simplicity of design, and insensitivity to toxic pollutants. Recently, clay and its modified forms have been used as adsorbents, and there has been an upsurge of interest in the interactions between dyes and clay particles. Clay may serve as an ideal adsorbent because of its low cost. It has relatively large specific surface area, excellent physical and chemical stability, and other advantageous structural and surface properties. Use of clay (especially three-layer clays) as adsorbent has been elaborately presented by Tolga Depci and Mehmet S. Çelik in Chapter 9.

Chapter 10 is about non-conventional adsorbents including clays, siliceous materials, zeolites, agricultural solid wastes, industrial byproducts, peat, chitin and chitosan, biomass, starch-based derivatives and miscellaneous adsorbents.Their effectiveness as an alternative green approach for the removal of dyes from wastewater and industrial effluents is discussed.

Hen feather is an abundantly available waste material found at poultry houses. It possesses marvelous and proficient structures, which are flexible as well as strong. Hen feather is composed of keratin and is biochemically similar to the substance responsible for creating the fur of mammals, scales of reptiles, horns of animals and fingernails of humans.

It is now well established that hen feather can be used as a potential adsorbent for the removal of hazardous pollutants. Before the year 2006, the use of hen feather as adsorbent was limited to the removal of metal ions only. However, in an innovative initiative first made by Alok Mittal and Jyoti Mittal, it was found that hen feather can also be exploited as a dye scavenger for wastewater. Chapter 11 summarizes the results of the removal of dye contaminants from water using hen feather as an adsorbent. The chapter provides comparable consequences of the effects of various parameters influencing the adsorption, various adsorption isotherms, kinetics, etc., of the developed dye removal processes.

The main outcome of reading this book will be that the reader is going to have a holistic view of the immense potential and ongoing research in dye removal by green chemistry, and its close connection with modern research and engineering applications. Furthermore, this book can be used as an important platform to inspire researchers in any related fields to develop greener processes for important techniques for use in several fields.

I gratefully acknowledge all the contributors of this book, without whom these valuable chapters could not have been completed. I express my highest gratitude and thankfulness to all of them.

Sanjay K. Sharma, FRSC
Jaipur, India
1st January 2015

Acknowledgements

When you complete a task and take time to rewind your journey and relive it through memories, you find some smiling and encouraging faces that have motivated you to complete the task with untiring efforts to your full ability. Such smiling faces remove the pain of stress which we occasionally face during any journey and encourage us to "Go ahead." They deserve a special mention and gratitude, love and affection.

It is time for me to express my feelings about my friends, colleagues, supporters and well- wishers and to let them know that I was so fortunate to have them and their valuable cooperation during the writing of this book, *Green Chemistry for Dye Removal from Wastewater: Research Trends and Applications*.

First of all I want to express my special thanks to all esteemed contributors of this book, who deserve special mention for contributing their writings, without which this book would not have been possible.

I deeply acknowledge my parents, Dr. M.P. Sharma and Mrs. Parmeshwari Devi, for their never-ending encouragement, moral support and blessings.

My wife Dr. Pratima Sharma deserves the highest appreciation for being beside me all the way and encouraging me in every hour of crisis. I appreciate her patience over the course of this book.

I also wish to thank Mr. Amit Agarwal and Mr. Arpit Agarwal (Vice Chairpersons, JECRC University, Jaipur) for their never ending support and encouragement, Prof. Victor Gambhir (President, JECRC University, Jaipur), Prof. J.K. Sharma (Pro-President, JECRC University, Jaipur) , Prof. R.N. Prasad (Dean, School of Sciences) and Prof. D.P. Mishra (Registrar, JECRC University, Jaipur) for their appreciation and guidance.

My kids Kunal and Kritika always deserve special mention as they are my best companions, who energize me to work with a refreshed mood and renewed motivation.

Special thanks go to Martin and his team behind this publication, without whose painstaking efforts this work could not have been completed in a timely manner.

I am also thankful to many others whose names I have not been able to mention but whose association and support has not been less in any way.

About the Editor

Prof. (Dr.) Sanjay K. Sharma is a very well-known author and editor of many books, research journals and hundreds of articles over the last twenty years.

Presently Prof. Sharma is working as Professor and Head of the Department of Chemistry, JECRC University, Jaipur, India, where he is teaching Engineering Chemistry and Environmental Chemistry to B. Tech Students; Green Chemistry, Spectroscopy and Organic Chemistry to undergraduate and post-graduate students; and pursuing his research interest in the domain of Green Chemistry with special reference to Water Pollution, Corrosion Inhibition and Biopolymers.

Dr. Sharma has had 16 books published on Chemistry by national-international publishers and over 61 research papers of national and international repute to his credit.

He has also been appointed as a Series Editor by Springer, UK, for their prestigious book series "Green Chemistry for Sustainability," where he has been involved in editing 14 different titles by various international contributors so far. Dr. Sharma is also serving as Editor-in-Chief for the *RASAYAN Journal of Chemistry*

He is a Fellow of the Royal Society of Chemistry (UK), member of the American Chemical Society (USA), and International Society for Environmental Information Sciences (ISEIS, Canada) and is also a life-time member of various international professional societies including the

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Removal of Organic Dyes from Industrial Effluents: An Overview of Physical and Biotechnological Applications

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Abstract

The textile industry produces a large amount of dye effluents, which are highly toxic as they contain a large number of metal complex dyes. The use of synthetic chemical dyes in various industrial processes, including paper and pulp manufacturing, plastics, dyeing of cloth, leather treatment and printing has increased considerably over the last few years, resulting in the release of dye-containing industrial effluents into the soil and aquatic ecosystems. The textile industry generates highly polluted wastewater and its treatment is a very serious problem due to high total dissolved solids (TDS), the presence of toxic heavy metals and the non-biodegradable nature of the dyestuffs present in the effluents. There are many processes available for the removal of dyes by conventional treatment technologies including biological and chemical oxidation, coagulation and adsorption, but they cannot be effectively used individually.

Many approaches, including physical, chemical and/or biological processes have been used in the treatment of industrial wastewater containing dye, but such

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Sanjay K. Sharma (ed.) Green Chemistry for Dyes Removal from Wastewater, (1–34)
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methods are often very costly and not environmentally safe. Furthermore, the large amount of sludge generated and the low efficiency of treatment with respect to some dyes have limited their use.

Keywords: Natural dyes, acid dyes, disperse dyes, cationic dyes, adsorption, membrane filtration, ion exchange, irradiation, electrokinetic coagulation, aerobic and anaerobic degradation

1.1 Introduction

Water, which is one of the abundant compounds found in nature, covers approximately three-fourths of the surface of the earth. Over 97% of the total quantity of water is in the oceans and other saline bodies of water and is not readily available for our use. Over 2% is tied up in polar ice caps and glaciers and in atmosphere and as soil moisture. As an essential element for domestic, industrial and agricultural activities, only 0.62% of water found in fresh water lakes, rivers and groundwater supplies, which is irregularly and non-uniformly distributed over the vast area of the globe, is accessible [1].

A reevaluation of the issue of environmental pollution made at the end of the last century has shown that wastes such as medicines, disinfectants, contrast media, laundry detergents, surfactants, pesticides, dyes, paints, preservatives, food additives, and personal care products which have been released by chemical and pharmaceutical industries, are a severe threat to the environment and human health on a global scale [2]. The progressive accumulation of more and more organic compounds in natural waters is mostly a result of the development of chemical technologies towards organic synthesis and processing. The population explosion and expansion of urban areas have had an increased adverse impact on water resources, particularly in regions in which natural resources are still limited. Currently, water use or reuse is a major concern which needs a solution. Population growth leads to a significant increase in default volumes of wastewater, which makes it an urgent imperative to develop effective and low-cost technologies for wastewater treatment [3].

Especially in the textile industry, effluents contain large amounts of dye chemicals which may cause severe water pollution. Also, organic dyes are commonly used in a wide range of industrial applications. Therefore, it is very important to reduce the dye concentration of wastewater before discharging it into the environment. Discharging large amounts of dyes into water resources, organics, bleaches, and salts, can affect the physical and chemical properties of fresh water. Dyes in wastewater that can obstruct light penetration and are highly visible, are stable to light irradiation and heat and also toxic to microorganisms. The removal of dyes is a very complex process due to their structure and synthetic origins [4].

Dyes that interfere directly or indirectly in the growth of aquatic organisms are considered hazardous in terms of the environment. Nowadays a growing awareness has emerged on the impact of these contaminants on ground water, rivers, and lakes [5–8].

The utilization of wastewater for irrigation is an effective way to dispose of wastewater [9]. Although various wastewater treatment methods including physical, chemical, and physicochemical have been studied, in recent years a wide range of studies have focused on biological methods with some microorganisms such as fungi, bacteria and algae [10]. The application of microorganisms for dye wastewater removal offers considerable advantages which are the relatively low cost of the process, its environmental friendliness, the production of less secondary sludge and completely mineralized end products which are not toxic [11]. Numerous researches on dye wastewater removal have been conducted which have proven the potential of microorganisms such as *Cunninghamella elegans* [12], *Aspergillus nigerus* [13], *Bacillus cereus* [14], *Chlorella sp.* [15] and also *Citrobacter sp*. [16,17].

1.1.1 Dyes

A dye or a dyestuff is usually a colored organic compound or mixture that may be used for imparting color to a substrate such as cloth, paper, plastic or leather in a reasonably permanent fashion. The dye that is generally described as a colored substance should have an affinity for the substrate or should fix itself on the substrate to give it a permanent colored appearance, but all the colored substances are not the dye [18,19]. Unlike many organic compounds, the dyes which contain at least one chromophore group and also a conjugated system and absorb light in the visible spectrum (400–700 nm) and exhibit the resonance of electrons, possess special colors [20].

The relationships between wavelength of visible and color absorbed/observed [21] are given on Table 1.1.

In general, a small amount of dyes in aqueous solution can produce a vivid color because they have high molar extinction coefficients. Color can be quantified by spectrophotometry (visible spectra), chromatography (usually high performance liquid, HPLC) and high performance capillary electrophoresis [19].

Table 1.1 Wavelengths of light absorption versus the color of organic dyes.

Wavelength range absorbed (nm)	Color absorbed	Color observed
400–435	Violet	Yellow-Green
435–480	Blue	Yellow
480–490	Green-Blue	Orange
490–500	Blue-Green	Red
500–560	Green	Purple
560–580	Yellow-Green	Violet
580–595	Yellow	Blue
595–605	Orange	Green-Blue
605–700	Red	Blue-Green

With regard to their solubility, organic colorants fall into two classes, dyes and pigments. The key distinction is that dyes are soluble in water and/or an organic solvent, while pigments are insoluble in both types of liquid media. Dyes are used to color substrates to which they have a specific affinity, whereas pigments can be used to color any polymeric substrate by a mechanism quite different than that of dyes [22,21].

1.1.2 Historical Development of Dyes

Humans discovered that certain roots, leaves, or bark could be manipulated, usually into a liquid form, and then used to dye textiles. They used these techniques to decorate clothing, utensils, and even the body, as a religious and functional practice. Records and cloth fragments dating back over 5000 years ago indicate intricate dyeing practices. Certain hues have historical importance and denote social standing [23]. The dye made from the secretions of shellfish, which is a clear fluid that oxidizes when exposed to the air, was used to produce a red to bluish purple. This dye was difficult to create and used only on the finest garments; hence it became associated with aristocrats and royalty [23]. Until the middle of the last century most of the dyes were derived from plants or animal sources by long and elaborate processes. Ancient Egyptian hieroglyphs contain a thorough description of the extraction of natural dyes and their application in dyeing [18]. In the past, only organic matter was available for use in making dyes. Today, there are numerous options and methods for the colorization of textiles. While today's methods capitalize on efficiency, there is question as to whether the use of chemicals is harmful to the environment.

In 1856, Sir William Perkin discovered a dye for the color mauve, which was the first synthetic dye. The method related to the dyeing of this color using coal and tar led to many scientific advances and the development of synthetic dyes [24,25].

Initially the dye industry was based on the discovery of the principal that dye chromogens associated with a basic arrangement of atoms were responsible for the color of a dye. Essentially, apart from one or two notable exceptions, all the dye types used today were discovered in the 1800s. The discovery of reactive dyes in 1954 and their commercial launch in 1956 heralded a major breakthrough in the dyeing of cotton; intensive research into reactive dyes followed over the next two decades and, indeed, is still continuing today. The oil crisis in the early 1970s, which resulted in a steep increase in the prices of dyestuff, created a driving force for more low-cost dyes, both by improving the efficiency of the manufacturing processes and by replacing tinctorially weak chromogens, such as anthraquinone, with tinctorially stronger chromogens, such as (heterocyclic) azo and benzodifuranone [26,27].

1.1.3 Natural Dyes

Natural dyes which are obtained from plants, insects/animals and minerals are renewable and sustainable bioresource products with minimum environmental impact. They have been known since antiquity for their use in coloring of textiles, food substrate, natural protein fibers like wool, silk and cotton, and leather as well as food ingredients and cosmetics [28–32].

Also, natural dyes are known for their use in dye-sensitized solar cells [33], histological staining [34], as a pH indicator [35] and for several other disciplines [36,37].

Over the last few decades, there has been increasing attention on various aspects of natural dye applications, and extensive research and development activities in this area are underway worldwide [29].

1.2 Classification of Dyes

Dyes may be classified according to their chemical structures and their usage or application methods. Dyes have different chemical structures derived from aromatic and hetero-aromatic compounds, and their chromophor and auxochrom groups mainly differ [18].

The most appropriate system for the classification of dyes is by chemical structure, which has many advantages. First, it readily identifies dyes as belonging to a group that has characteristic properties, for example, azo dyes (strong, good all-round properties, low-cost) and anthraquinone dyes (weak, expensive). Second, there are a number of manageable chemical groups. Most importantly, it is the classification used most widely by both the synthetic dye chemist and technologist. Thus, both chemists and technologists can readily identify with phrases such as an azo yellow, an anthraquinone red, and a phthalocyanine blue [27].

The application classification of dyes arranged according to the C. I. (Color Index) is given in Table 1.2, which includes the principal substrates, the methods of application, and the representative chemical types for each application class [27]. Although not shown in Table 1.2, dyes are also used in high-tech applications, such as in medical, electronics, and especially the nonimpact printing technologies [27,38].

Acid Dyes, which are water-soluble anionic dyes, are applied to nylon, wool, silk and modified acrylics. They are also used to some extent for paper, leather, inkjet printing, food, and cosmetics.

Direct Dyes are water-soluble anionic dyes. When dyed from aqueous solution in the presence of electrolytes, they are substantive to, i.e., have high affinities for cellulosic fibers. Their principal use is in the dyeing of cotton and regenerated cellulose, paper, leather, and, to a lesser extent, nylon. Most of the dyes in this class are polyazo compounds, along with some stilbenes, phthalocyanines, and oxazines. Treatments applied to the dyed material to improve wash fastness properties include chelation with salts of metals, which are usually copper or chromium, and treatment with formaldehyde or a cationic dye-complexing resin.

Azoic Dyes are applied via combining two soluble components impregnated in the fiber to form an insoluble color molecule. These dye components, which are sold as paste-type dispersions and powders, are chiefly used for cellulosic fibers, especially cotton. Dye bath temperatures of 16–27°C (60–80°F) are generally used to make the shade [39].

Disperse Dyes, which are substantially water-insoluble nonionic dyes for application to hydrophobic fibers from aqueous dispersion, are used predominantly on polyester and to a lesser extent on nylon, cellulose, cellulose acetate, and acrylic fibers. Thermal transfer printing and dye diffusion thermal transfer (D_2T_2) processes for electronic photography represent rich markets for selected members of this class.

Sulfur Dyes are used primarily for cotton and rayon. The application of sulfur dyes requires carefully planned transformations between the water-soluble reduced state of the dye and the insoluble oxidized form. Sulfur dyes, which generally have a poor resistance to chlorine, and are not applicable to wool or