Colorants in Water and Toxicity

Subjects: Environmental Sciences

Contributor: Prithvi Srivatsav

Dyes (colorants) are used in many industrial applications, and have become vital to the industrial production infrastructure. However, effluents of some dyeing industries contain toxic chemicals, which are detrimental to both the environment and human health. Therefore, wastewater containing dyes must be properly treated before discharging to surrounding water bodies, and this paper summarises that the most effective current method of effluent treatment for dye industries is adsorption using Biochar.

adsorption biochar dyes removal wastewater treatment

1. Introduction

Water pollution has become a major environmental problem globally. Different types of contaminants, mainly discharged from the industrial and agricultural activities, significantly contribute to water pollution. One of the major contributors to water pollution is the release of untreated dye effluents. These dyes are not only harmful to the plants and aquatic life, but also to human beings. Cancer, allergies, skin diseases are some of the health complications that may arise due to the ingestion and absorption of dye-contaminated water by humans [1]. Biochar has been identified as a potential candidate for wastewater treatment. The use of biochar for wastewater treatment has an added advantage due to the availability of abundance of surface functional groups on biochar surface and having a large surface area. Biochar is produced by the decomposition of biomass by thermochemical methods in the absence or low amount of oxygen. Different biomass feedstocks could be utilized to synthesize biochar, which include algae, crop residues, forests biomass, and animal manures. Torrefaction, hydrothermal carbonization, microwave heating, and gasification are some of the methods available for thermal decomposition of biomass [2][3]. Before biochar can be used, it has to be subjected to pretreatment processes (e.g., sieving, washing, crushing, etc.), followed by pyrolysis.

2. Colorants in Water and Toxicity

2.1. Natural and Synthetic Dyes

Dyes can be defined as color-imparting organic compounds that are water or oil soluble; they are distinguished from pigments that are insoluble. Natural dyes are those that are derived or extracted from natural sources, such as animals, flowers, roots, mollusks, minerals, etc. Natural dyes are broadly classified into two types: adjective and substantive [4]. Adjective dyes give permanent color only when used along with a mordant to bind them to the fabric; meanwhile substantive dyes contain a natural mordant, called tannin, and can give fast color without the use of additional mordants [5]. Mordants are compounds that act as a bridge between the fabric molecules and the dye, holding it in place to promote fastness [6]. Weak acids, such as tannic acid and acetic acid, are some natural mordants [7][8], while metal salts, such as copper sulfate and ferrous sulfate, can also be utilized. The use of natural dyes is advantageous as these are relatively non-toxic and renewable in nature, but their use is not cost-effective in the industrial context, and they fail to give an even consistent hue when compared to the synthetic dyes. Natural dyes may still be used in domestic and small-scale operations.

Synthetic dyes are usually unsaturated organic molecules. The first synthetic dye prepared was Mauve, a reddish-purple dye, which quickly degraded under water or direct sunlight to form a pale purple color. The first synthetic dye was derived from coal tar, the product resulting from the carbonization of coal. In the current context, synthetic dyes are more economical and exhibit better color fastness in comparison to natural dyes; thus, they dominate the market. Though they are more economical, synthetic dyes are also decidedly more toxic and polluting than natural dyes, causing environmental pollution and adverse health effects on living organisms [5].

2.2. Classification of Dyes

2.2.1. Chromophores and Color Index (C.I.)

Atoms or groups of atoms are said to be chromogenic if they are able to impart colour to the dye, and different arrangements and numbers of these chromogenic groups in a molecule impart different colors to the dyes. The other atoms or groups of atoms in the molecule, which are bound to the chromophore, and influence or bring a change in the color of the dye, are called auxochromes [9]. Thus, different textile dyes have specific structures that contribute to their characteristic colors. Some examples of chromophores are carbonyl groups, nitro groups, azo groups, or a conjugated pi electron system. Groups such as hydroxyl, aniline, and sulfonic acid act as auxochromes [10]. Dyes are classified by their applications and are given a unique identifying name called the Color Index (CI). As dye structures can be very complex, referring to it by its chemical name can be impractical, and common names can change between regions; thus, the name of the dye is standardized by using its CI. Every dye is issued a generic name and a CI number; the generic name consists of the method of application of the dye (e.g., direct, reactive etc.), the hue, and an identification number. The dye is also given a CI number based on the functional group and configuration of the molecule. For example, the dye commonly known as Remazol Red B is referred to as CI Reactive Red 22 (CI number: 14,824) under CI classification rules [11].

2.2.2. Disperse Dyes

Disperse dyes are mostly non-ionic in nature and are insoluble or very sparingly soluble in water; they are named so because their application involves dispersing the dye into a very fine suspension in solvent [12]. The chromogenic groups in disperse dyes are azo, anthraquinone, or nitro groups. The non-ionic nature of disperse dyes make them an ideal choice for dying synthetic and hydrophobic fibers, such as acetate, polyester, and sometimes nylon and acrylic fibers [13]. This is because these fibers are negatively charged and so reactive or basic dyes cannot be used. Only the non-ionic disperse dyes, which are not affected by surface charge, are able to reliably dye these fabrics. Traditionally, a carrier is added to the dye to improve the dispersion action and to aid dyeing. Disperse dyes can be classified from class A to class D based on their sublimation temperature, with class A having the lowest and class D the highest [14]. Since sublimation temperature depends on the size of the molecule, class A dyes have lower relative molecular size compared to class D dyes. Due to its sparing solubility, non-ionic nature, and non-biodegradability, disperse dyes are particularly harmful when left untreated in wastewater [15].

2.2.3. Direct Dyes

Direct dyes are water-soluble, anionic dyes that, unlike reactive or vat dyes, can be applied directly onto substrates using a neutral or alkaline bath (using sodium chloride or sodium sulfate) [16]. These dyes have an affinity to cellulosic materials, such as paper, cardboard, cotton, etc., but can also be applied on fabrics, such as, rayon and silk, with the use of mordants. The chromogenic functional groups in direct dyes include stilbene, phthalocyanine, oxazine, thiazole, but mainly azo groups [17]. Direct dyes are advantageous compared to other dyes because they have superior lightfastness compared to most reactive dyed hues, even though they cost less and are, thus, more economical. They also require less water use and the salt concentrations of the effluent are much less, compared to most reactive dyes. Some direct dyes, such as, Congo red, are carcinogenic, and have been banned from use [18].

2.2.4. Reactive Dyes

Reactive dyes have become one of the most widely used synthetic dyes in the industrial context due to their excellent wash fastness and bright and varied types of hues [19]. The chromophores in reactive dyes are largely azo groups; blue and green colors are given due to the presence of anthraquinone and phthalocyanine structures [20]. Direct dyes are largely used to dye cellulosic substrates and fibers, although other substrates can be used too. Unlike direct dyes, however, reactive dyes form new covalent bonds with the nucleophilic sites in fabric molecules, thus leading to its remarkable wash fastness [11]. Some of the most commonly utilized groups in reactive dyes are trichloropyrimidine, sulphatoethylsulphone, dichloroquinoxaline, and dichlorotriazin [21]. The major disadvantage regarding reactive dyes is their environmental threat. Effluent from dyeing cotton, using reactive dyes, are extremely polluted, having very high chemical oxygen demand (COD), salt load, and visible color in water [22]. Both, the unfixed dye and its hydrolyzed form, are soluble in water, and thus their removal is particularly challenging [23]. Some reactive dyes are also associated with heavy metals, such as chromium, copper, or nickel, and these can later be released into aquatic ecosystems on degradation of the dye molecule.

2.3. Toxicity of Dyes

The global textile industry is estimated to be worth around $1 trillion USD and its contribution towards total world exports is around 7%, employing 35 million people worldwide [24]. Thus, this industry has a high impact on the environment and human health in general, due to the pollution it causes. The most prominent and destructive form of pollution, caused by the textile industry, is the water pollution due to manufacturing of dyes. Textile effluents are both aesthetically polluted, and have high salinity, chemical oxygen demand, and ecotoxicity [25], and due to their increasing ubiquity in surface water, can lead to adverse effects to human and wildlife health and to aquatic ecosystems in general. Most synthetic dyes are highly toxic to humans and aquatic beings, and have acute and chronic effects. For example, reactive dyes are notorious, causing health issues such as, dermatitis, occupational asthma, rhinitis, and other allergic reactions for the workers involved in these dyes manufacturing [23]. Dyes are also mutagenic and carcinogenic in nature [26][27], which leads to chronic effects, such as kidney, urinary bladder, and liver cancer in dye workers. A xanthene dye called erythrosine is carcinogenic, neurotoxic and DNA-damaging for humans and animals alike [20]. Metal complexed dyes, which are widely used for their resistance, have heavy metals, such as copper, nickel, and chromium. When discharged to aquatic environments, these metals can be taken up by fish gills and can be transferred to humans through the food chain [27]. Current treatment methods are inadequate to treat dye effluents effectively, because of their recalcitrant nature in aerobic environments [28], and thus, these substances can linger in soil and lead to bioaccumulation, leading to complications in organisms higher up the food chain [29]. Thus, current effluent treatment techniques are inadequate for the dyeing industry and to prevent the further insemination of surface water with such mutagenic and carcinogenic molecules, we must adopt novel and more effective treatment techniques, such as bioremediation or biochar adsorption.

3. Treatment Technologies for Dyes Removal from Water

3.1. Coagulation

Coagulation is one of the most popular wastewater treatment techniques used since the early 20th century. Coagulation is the process of adding chemical compounds to bind particles together until they acquire a large mass to ultimately settle down. These chemicals are known as coagulants and carry a positive charge, which are mixed rapidly in the wastewater for uniform distribution. Most of the dissolved/suspended particles encountered in wastewater carry a negative charge, which are neutralized by these coagulants, thus making these capable of sticking together. Coagulation process is usually employed as a preliminary step in wastewater treatment process. The most frequently used coagulants are iron or aluminum salts. However, wastewater (containing dyes) is rich in color, and have high COD levels. Hence, conventional methods, such as coagulation, prove inefficient and moreover cause the problem of sludge disposal [30]. A combination of the conventional coagulation technique with other treatment methods must be innovated to improve its efficiency. For example, combination of coagulation and adsorption techniques was a feasible way for the removal of reactive dyes from water [31]. The adsorbent used was activated carbon, derived from coconut shells, and the coagulation process was carried out using aluminum chloride as the coagulant. It was found that the removal efficiencies for Orange 16 and Black 5 reactive dyes were 84% and 90%, respectively [31].

3.2. Advanced Oxidation Processes (AOPs)

AOPs are chemical treatment methods used to remove the organic/inorganic contaminants present in wastewater by the oxidizing action caused due to the in-situ production of hydroxyl radicals ($^{\bullet}OH$) [32]. These radicals are produced by oxidizing agents (H_2O_2, O_3, $KMnO_4$), catalysts, or UV light. Some of the AOPs include photocatalysis, ozonation etc. [33]. The following sections briefly explain some of the AOPs used in dye removal from wastewater.

3.2.1. Ozonation

Most of the dyes, encountered in wastewater, have polycyclic aromatic structures containing elements, such as nitrogen, metals, and sulfur, which makes it difficult to treat the wastewater by physical, chemical, and biological methods. Conjugated chains present in the dye structures, responsible for imparting color, are actively destroyed by ozonation, which is an AOP method that involves the chemical treatment of wastewater by dissolving ozone in water [34]. Employing ozonation as a treatment technique is advantageous as zero sludge is generated. Degradation of the dye is achieved in a single step and furthermore, ozone decomposes into stable oxygen [35]. However, ozonation as a treatment process very rarely results in complete oxidation, so by-products are usually formed in the effluent stream [36].

3.2.2. Fenton's Reagent and Fenton-Like Processes

Fenton's reagent is a solution of ferrous iron along with hydrogen peroxide, which finds its use as a catalyst in oxidizing various contaminants present in wastewater. A composite of La-Fe-O [37] was used as a photo-Fenton catalyst in the presence of light irradiation and hydrogen peroxide, and it was found that rhodamine B dye was oxidized to 98% within 25 min.

3.3. Membrane Processes

Membranes have been in use for wastewater treatment as early as the 1960s. Since membrane processes were too expensive at that time, this process was only chosen for specialized applications. Since the 2000s, membranes have been made cost-effective and are being used with other conventional water treatment processes. A membrane is a thin, semi-permeable material that is attached to a porous support, and is used for the removal of dissolved substances, based on properties, such as, size or charge, when a driving force is applied on it. Membrane processes are used in reverse osmosis (RO), forward osmosis, nanofiltration, and ultrafiltration.

3.3.1. Nanofiltration

The membranes used for nanofiltration have pore size within the range of 0.1–10 nm. Nanofiltration membranes have the advantages of separating dyes with a high molecular weight and also have >90% rejection efficiency for dye removal, making it a promising approach towards dye removal [38]. For example, positively charged polyethylenimine-modified nanofiltration membrane showed semi-xylenol orange, Tropaeolin O, Victoria blue B dyes removal efficiency of 99%, 98.3%, and 99.2%, respectively [39].

3.3.2. Forward Osmosis (FO)

FO is a water treatment process that utilizes the osmotic pressure gradient to separate water from its dissolved solutes. Since FO process is completely devoid for the requirement of a driving force, it is more energy efficient compared to reverse osmosis and other membrane separation processes [40]. A thin-film composite membrane was used under forward osmosis by [41], and it was found that it had a dye rejection rate of ≥96% for commonly used dyes in the textile industries.

3.4. Biological Process

Biological process involves the usage of bacteria, microbes, and other microorganisms to treat wastewater. These biological processes are environmentally friendly, energy saving, generate low sludge, and require zero to minimal amount of chemicals to be used. The efficiency of biological processes could further be increased by varying the environmental conditions to favor the growth of the microorganisms. Algae is widely used by researchers as a potential option for the purpose of biosorption, as algae contains proteins, lipids and functional groups such as amino, carboxylate, sulfate, etc., in its structure [42]. Since algae possesses a wide surface area and excellent binding affinity in its cell structure, it results in high biosorption capabilities [43]. As algae is easily accessible (highly abundant in saltwater oceans and freshwater lakes), the use of algae could subsequently be extended for the dye removal from textile wastewater. For example, chemically (sulfuric acid) modified defatted Laminaria japonica biomass (renewable brown algae) showed methylene blue adsorption capacity of 549.45 mg/g, and quasi-equilibrium was achieved within 60 min under optimal conditions with a biosorbent dose of 0.6 g/L, pH 6 and temperature of 308 K [44].

Bioreactors are also becoming increasingly popular for dyes removal. Usually, a combination of both aerobic and anaerobic processes is used for the treatment of azo dyes. For example, the removal of Alizarin Yellow R dye was studied by the combined process of up-flow bio-electrocatalyzed electrolysis reactor and aerobic bio-contact oxidation reactor in just 6 h of hydraulic retention time [45]. Moreover, the COD removal efficiency and decolorization efficiency was found to be 93.0 ± 0.5% and 93.8 ± 0.7%, respectively, in the process.

3.5. Adsorption Process

As far as dye removal from water is concerned, adsorption has been found as one of the best treatment processes among other conventional water treatment methods due to its low-cost, affordability, greater efficiency, and the fact that it requires minimum maintenance [46]. Adsorption also has the added advantage of producing no detrimental residues and having the capacity to treat a large volumes of water [47]. An adsorbent could also be recycled multiple times for its usage in subsequent treatment processes [48]. Common materials, such as activated carbon, zeolites, activated alumina, silica gel and polymeric adsorbents have been widely used for water treatment. The use of biomaterials for adsorption processes instead of conventional materials is now being the subject of interest by many researchers as the commercial value of biomaterials is low, and also they are available in abundance [49]. Naturally-derived biopolymers, which are hyperreactive, chemically stable, possess good physicochemical properties have garnered significant attention that is worth looking into for employing these biopolymers for the role of green adsorbents [50]. Table 1 highlights the major advantages and disadvantages of treatment techniques, available for dye removal from water.

Table 1. The advantages and disadvantages of various treatment technologies for dyes removal from water.

Treatment Technology	Advantages	Disadvantages

for Dyes Removal		
Coagulation	· Reduced time for settling of suspended solids · Easy removal of fine particles · Effective in removing bacteria, protozoa, and virus	· High cost to spend for frequent monitoring and accurate dosing · Huge volume of sludge generation
Advanced Oxidation Processes (AOP)	· •OH radicals can treat a wide range of organic material · Zero sludge production	· Fenton's reagent AOP results in iron sludge generation · High capital and maintenance costs are expected
Membrane Processes	· Minor, valuable products can be recovered from the feed stream · Process can be easily "scaled-up" · No-phase changes involve between the feed and product stream · Eco-friendly as simple and non-toxic materials are used	· High flow rate has the potential to damage the membrane · This process results in membrane fouling effects. Regeneration and extensive cleaning are required. · High equipment cost
Biological Processes	· Almost all biodegradable organic matter is effectively removed · Efficient attenuation of color · Eco-friendly and a common wastewater treatment mechanism	· Slow process · An optimal favorable environment is crucial · Biological sludge generation · Remediation of dye molecules is tough

Adsorption Processes	· Highly efficient process · Applicable for a wide variety of target contaminants · Treatment technology is easy to employ	· Deterioration of adsorbent performance when subjected to multiple operational cycles · Spent adsorbent is likely to be a hazardous waste · Regeneration of adsorbent material is expensive

source:
.12,

2. Kazemi Shariat Panahi, H.; Dehhaghi, M.; Ok, Y.S.; Nizami, A.S.; Khoshnevisan, B.; Mussatto, S.I.; Aghbashlo, M.; Tabatabaei, M.; Lam, S.S. A comprehensive review of engineered biochar: Production, characteristics, and environmental applications. J. Clean. Prod. 2020, 270, 122462, doi:10.1016/j.jclepro.2020.122462.
3. Chi, N.T.L.; Anto, S.; Ahamed, T.S.; Kumar, S.S.; Shanmugam, S.; Samuel, M.S.; Mathimani, T.; Brindhadevi, K.; Pugazhendhi, A. A review on biochar production techniques and biochar based catalyst for biofuel production from algae. Fuel 2020, 119411, doi:10.1016/j.fuel.2020.119411.
4. Bhuiyan, M.A.R.; Islam, A.; Ali, A.; Islam, M.N. Color and chemical constitution of natural dye henna (Lawsonia inermis L) and its application in the coloration of textiles. J. Clean. Prod. 2017, 167, 14–22, doi:https://doi.org/10.1016/j.jclepro.2017.08.142.
5. Zerin, I.; Farzana, N.; Sayem, A.S.M.; Anang, D.M.; Haider, J. Potentials of Natural Dyes for Textile Applications; Hashmi, S., Choudhury, Eds.; Elsevier: Oxford, UK, 2020; pp. 873–883, ISBN 978-0-12-813196-1.
6. Nambela, L.; Haule, L.V.; Mgani, Q. A review on source, chemistry, green synthesis and application of textile colorants. J. Clean. Prod. 2020, 246, 119036, doi:https://doi.org/10.1016/j.jclepro.2019.119036.
7. Saxena, S.; Raja, A.S.M. Natural Dyes: Sources, Chemistry, Application and Sustainability Issues BT - Roadmap to Sustainable Textiles and Clothing: Eco-friendly Raw Materials, Technologies, and Processing Methods. In; Muthu, S.S., Ed.; Springer Singapore: Singapore, 2014; pp. 37–80 ISBN 978-981-287-065-0.
8. Degano, I.; Mattonai, M.; Sabatini, F.; Colombini, M.P. A mass spectrometric study on tannin degradation within dyed woolen yarns. Molecules 2019, 24, doi:10.3390/molecules24122318.
9. Christie, R. Colour Chemistry; Royal Society of Chemistry: Cambridge, UK, 2014; ISBN 1849733287.
10. Raman, C.D.; Kanmani, S. Textile dye degradation using nano zero valent iron: A review. J. Environ. Manage. 2016, 177, 341–355, doi:https://doi.org/10.1016/j.jenvman.2016.04.034.

11. Gürses, A.; Açıkyıldız, M.; Güneş, K.; Gürses, M.S. Classification of Dye and Pigments BT - Dyes and Pigments. In; Gürses, A., Açıkyıldız, M., Güneş, K., Gürses, M.S., Eds.; Springer International Publishing: Cham, 2016; pp. 31–45 ISBN 978-3-319-33892-7.
12. Clark, M. Fundamental principles of dyeing. Handb. Text. Ind. Dye. M. Clark (ed). Woodhead Publ. Ser. Text. 2011, 1, 1–27.
13. Burkinshaw, S.M. Physico-Chemical Aspects of Textile Coloration; SDC-Society of Dyers and Colourists; Wiley: Hoboken, USA, 2015; ISBN 9781118725634.
14. Roy Choudhury, A.K. 2—Dyeing of synthetic fibres. In Woodhead Publishing Series in Textiles; Clark, M. Eds.; Woodhead Publishing: Cambridge, UK, 2011; Volume 2, pp. 40–128, ISBN 978-1-84569-696-2.
15. Jamil, A.; Bokhari, T.H.; Javed, T.; Mustafa, R.; Sajid, M.; Noreen, S.; Zuber, M.; Nazir, A.; Iqbal, M.; Jilani, M.I. Photocatalytic degradation of disperse dye Violet-26 using TiO2 and ZnO nanomaterials and process variable optimization. J. Mater. Res. Technol. 2020, 9, 1119–1128, doi:https://doi.org/10.1016/j.jmrt.2019.11.035.
16. Broadbent, A.. Basic principles of textile coloration. Color Res. Appl. 2003, 28, 230–231, doi:10.1002/col.10152.
17. Chattopadhyay, D.P. 4—Chemistry of dyeing. In Woodhead Publishing Series in Textiles; Clark, M. Eds.; Woodhead Publishing: Cambridge, UK, 2011; Volume 1, pp. 150–183, ISBN 978-1-84569-695-5.
18. Jalandoni-Buan, A.C.; Decena-Soliven, A.L.A.; Cao, E.P.; Barraquio, V.L.; Barraquio, W.L. Characterization and identification of Congo red decolorizing bacteria from monocultures and consortia. Philipp. J. Sci. 2010, 139, 71–78.
19. Pereira, L.; Alves, M. Dyes—Environmental Impact and Remediation BT - Environmental Protection Strategies for Sustainable Development. In; Malik, A., Grohmann, E., Eds.; Springer Netherlands: Dordrecht, 2012; pp. 111–162 ISBN 978-94-007-1591-2.
20. Pal, P. Chapter 6—Industry-Specific Water Treatment: Case Studies; Pal, P, Ed.; Butterworth-Heinemann: Oxford, UK, 2017; pp. 243–511, ISBN 978-0-12-810391-3.
21. Benkhaya, S.; M' rabet, S.; El Harfi, A. A review on classifications, recent synthesis and applications of textile dyes. Inorg. Chem. Commun. 2020, 115, 107891, doi:10.1016/j.inoche.2020.107891.
22. Khatri, A.; Peerzada, M.H.; Mohsin, M.; White, M. A review on developments in dyeing cotton fabrics with reactive dyes for reducing effluent pollution. J. Clean. Prod. 2015, 87, 50–57, doi:10.1016/j.jclepro.2014.09.017.

23. Chavan, R.B. 16—Environmentally friendly dyes. In Woodhead Publishing Series in Textiles; Clark, M, Eds.; Woodhead Publishing: Cambridge, UK, 2011; Volume 1, pp. 515–561, ISBN 978-1-84569-695-5.
24. Desore, A.; Narula, S.A. An overview on corporate response towards sustainability issues in textile industry. Environ. Dev. Sustain. 2018, 20, 1439–1459, doi:10.1007/s10668-017-9949-1.
25. Liang, Z.; Wang, J.; Zhang, Y.; Han, C.; Ma, S.; Chen, J.; Li, G.; An, T. Removal of volatile organic compounds (VOCs) emitted from a textile dyeing wastewater treatment plant and the attenuation of respiratory health risks using a pilot-scale biofilter. J. Clean. Prod. 2020, 253, 120019, doi:https://doi.org/10.1016/j.jclepro.2020.120019.
26. Khatri, J.; Nidheesh, P. V; Anantha Singh, T.S.; Suresh Kumar, M. Advanced oxidation processes based on zero-valent aluminium for treating textile wastewater. Chem. Eng. J. 2018, 348, 67–73, doi:https://doi.org/10.1016/j.cej.2018.04.074.
27. Lellis, B.; Fávaro-Polonio, C.Z.; Pamphile, J.A.; Polonio, J.C. Effects of textile dyes on health and the environment and bioremediation potential of living organisms. Biotechnol. Res. Innov. 2019, 3, 275–290, doi:10.1016/j.biori.2019.09.001.
28. Vikrant, K.; Giri, B.S.; Raza, N.; Roy, K.; Kim, K.-H.; Rai, B.N.; Singh, R.S. Recent advancements in bioremediation of dye: Current status and challenges. Bioresour. Technol. 2018, 253, 355–367, doi:https://doi.org/10.1016/j.biortech.2018.01.029.
29. Xiang, X.; Chen, X.; Dai, R.; Luo, Y.; Ma, P.; Ni, S.; Ma, C. Anaerobic digestion of recalcitrant textile dyeing sludge with alternative pretreatment strategies. Bioresour. Technol. 2016, 222, 252–260, doi:https://doi.org/10.1016/j.biortech.2016.09.098.
30. Issa Hamoud, H.; Finqueneisel, G.; Azambre, B. Removal of binary dyes mixtures with opposite and similar charges by adsorption, coagulation/flocculation and catalytic oxidation in the presence of CeO2/H2O2 Fenton-like system. J. Environ. Manage. 2017, 195, 195–207, doi:10.1016/j.jenvman.2016.07.067.
31. Furlan, F.R.; de Melo da Silva, L.G.; Morgado, A.F.; de Souza, A.A.U.; Guelli Ulson de Souza, S.M.A. Removal of reactive dyes from aqueous solutions using combined coagulation/flocculation and adsorption on activated carbon. Resour. Conserv. Recycl. 2010, 54, 283–290, doi:10.1016/j.resconrec.2009.09.001.
32. Babu, D.S.; Srivastava, V.; Nidheesh, P. V.; Kumar, M.S. Detoxification of water and wastewater by advanced oxidation processes. Sci. Total Environ. 2019, 696, 133961, doi:10.1016/j.scitotenv.2019.133961.
33. Nidheesh, P. V.; Zhou, M.; Oturan, M.A. An overview on the removal of synthetic dyes from water by electrochemical advanced oxidation processes. Chemosphere 2018, 197, 210–227, doi:10.1016/j.chemosphere.2017.12.195.

34. Colindres, P.; Yee-Madeira, H.; Reguera, E. Removal of Reactive Black 5 from aqueous solution by ozone for water reuse in textile dyeing processes. Desalination 2010, 258, 154–158, doi:10.1016/j.desal.2010.03.021.
35. Muniyasamy, A.; Sivaporul, G.; Gopinath, A.; Lakshmanan, R.; Altaee, A.; Achary, A.; Velayudhaperumal Chellam, P. Process development for the degradation of textile azo dyes (mono-, di-, poly-) by advanced oxidation process - Ozonation: Experimental & partial derivative modelling approach. J. Environ. Manage. 2020, 265, doi:10.1016/j.jenvman.2020.110397.
36. Mascolo, G.; Lopez, A.; Bozzi, A.; Tiravanti, G. By-products formation during the ozonation of the reactive dye uniblu-A. Ozone Sci. Eng. 2002, 24, 439–446, doi:10.1080/01919510208901633.
37. Xu, X.; Geng, A.; Yang, C.; Carabineiro, S.A.C.; Lv, K.; Zhu, J. One-pot synthesis of La – Fe – O @ CN composites as photo-Fenton catalysts for highly e ffi cient removal of organic dyes in wastewater. Ceram. Int. 2020, 46, 10740–10747, doi:10.1016/j.ceramint.2020.01.083.
38. Jin, J.; Du, X.; Yu, J.; Qin, S.; He, M.; Zhang, K.; Chen, G. High performance nanofiltration membrane based on SMA-PEI cross-linked coating for dye/salt separation. J. Memb. Sci. 2020, 611, 118307, doi:10.1016/j.memsci.2020.118307.
39. Qi, Y.; Zhu, L.; Shen, X.; Sotto, A.; Gao, C.; Shen, J. Polythyleneimine-modified original positive charged nano filtration membrane: Removal of heavy metal ions and dyes. Sep. Purif. Technol. 2019, 222, 117–124, doi:10.1016/j.seppur.2019.03.083.
40. Meng, L.; Wu, M.; Chen, H.; Xi, Y.; Huang, M.; Luo, X. Rejection of antimony in dyeing and printing wastewater by forward osmosis. Sci. Total. Environ. 2020, 745, doi:10.1016/j.scitotenv.2020.141015.
41. Lin, C.; Tung, K.; Lin, Y.; Dong, C.; Wu, C. l P re of. Process Saf. Environ. Prot. 2020, doi:10.1016/j.psep.2020.07.007.
42. Marzbali, M.H.; Mir, A.A.; Pazoki, M.; Pourjamshidian, R.; Tabeshnia, M. Removal of direct yellow 12 from aqueous solution by adsorption onto spirulina algae as a high-efficiency adsorbent. J. Environ. Chem. Eng. 2017, 5, 1946–1956, doi:10.1016/j.jece.2017.03.018.
43. Ihsanullah, I.; Jamal, A.; Ilyas, M.; Zubair, M.; Khan, G.; Atieh, M.A. Bioremediation of dyes: Current status and prospects. J. Water Process Eng. 2020, 38, 101680, doi:10.1016/j.jwpe.2020.101680.
44. Shao, H.; Li, Y.; Zheng, L.; Chen, T.; Liu, J. Removal of methylene blue by chemically modified defatted brown algae Laminaria japonica. J. Taiwan Inst. Chem. Eng. 2017, 80, 525–532, doi:10.1016/j.jtice.2017.08.023.
45. Cui, D.; Guo, Y.Q.; Lee, H.S.; Cheng, H.Y.; Liang, B.; Kong, F.Y.; Wang, Y.Z.; Huang, L.P.; Xu, M.Y.; Wang, A.J. Efficient azo dye removal in bioelectrochemical system and post-aerobic

bioreactor: Optimization and characterization. Chem. Eng. J. 2014, 243, 355–363, doi:10.1016/j.cej.2013.10.082.

46. Sirajudheen, P.; Karthikeyan, P.; Vigneshwaran, S.; Meenakshi, S. Synthesis and characterization of La(III) supported carboxymethylcellulose-clay composite for toxic dyes removal: Evaluation of adsorption kinetics, isotherms and thermodynamics. Int. J. Biol. Macromol. 2020, 161, 1117–1126, doi:10.1016/j.ijbiomac.2020.06.103.
47. Saxena, M.; Sharma, N.; Saxena, R. Highly efficient and rapid removal of a toxic dye: Adsorption kinetics, isotherm, and mechanism studies on functionalized multiwalled carbon nanotubes. Surfaces and Interfaces 2020, 21, 100639, doi:10.1016/j.surfin.2020.100639.
48. Morais da Silva, P.M.; Camparotto, N.G.; Grego Lira, K.T.; Franco Picone, C.S.; Prediger, P. Adsorptive removal of basic dye onto sustainable chitosan beads: Equilibrium, kinetics, stability, continuous-mode adsorption and mechanism. Sustain. Chem. Pharm. 2020, 18, doi:10.1016/j.scp.2020.100318.
49. Deniz, F. Adsorption Properties of Low-Cost Biomaterial Derived from Prunus amygdalus L. for Dye Removal from Water. Sci. World J. 2013, 2013, 961671, doi:10.1155/2013/961671.
50. Fan, H.; Ma, Y.; Wan, J.; Wang, Y.; Li, Z.; Chen, Y. Adsorption properties and mechanisms of novel biomaterials from banyan aerial roots via simple modification for ciprofloxacin removal. Sci. Total Environ. 2020, 708, 134630, doi:https://doi.org/10.1016/j.scitotenv.2019.134630.