Egyptian Journal of Chemistry
http://ejchem.journals.ekb.eg/

Application of marine algae separate and in combination with natural zeolite in dye adsorption from wastewater; A review

Ahmed Hamd[1,5*], Asmaa Ragab Dryaz[2], Mohamed Shaban[1,3], Hamad AlMohamadi[3,4], Sayed A. Ahmed[2], Refat El-sayed [6,7], N.K. Soliman[5]

[1]*Nanophotonics and Applications Lab, Physics Department, Faculty of Science, Beni-Suef University, Beni Suef 62514, Egypt*
[2] *Department of Chemistry, Faculty of Science, Beni-Suef University, Beni Suef 62511, Egypt*
[3]*Department of Physics, Faculty of Science, Islamic University in Almadinah Almonawara, Almadinah Almonawara, 42351, Saudi Arabia.*
[4]*Department of Chemical Engineering, Faculty of Engineering, Islamic University of Madinah, Madinah, Saudi Arabia*
[5] *Basic science department, faculty of oral and dental medicine, Nahda university, Beni-Suef , Egypt*
[6] *Department of Chemistry, University College in Al-Jamoum, Umm Al-Qura University, Makkah, Saudi Arabia.*
[7] *Chemistry Department, Faculty of Science, Benha University, Benha, Egypt*

Abstract

Industrial wastewater is obtained from industrial activities that include any solids that become useless during the manufacturing process. These wastes are considered as international green problems so significant solutions must be taken to confront these problems and reduce their environmental burden and effect. Removal of dyes from industrial wastes using zeolite and marine algae by adsorption technique has a lot of advantages as the zero cost, obtainability, high effectiveness, and ecological alternative source. Dyes adsorption onto zeolite and marine algae surfaces is a complicated method that affected by numerous factors like contact time, initial dye concentration, solution pH, catalyst weight and temperature. In this review, we present definition of marine algae and their classification, definition of Zeolite , industrial wastewater and their effect on the eco system, water treatment methods which include chemical, biological, Combinatorial method, nanotechnology-based and physical methods, uses of algae and natural zeolite in wastewater treatment and finally we discuss the factors that affect dyes adsorption onto zeolite and marine algae surfaces, such as contact time, temperature, solution pH and catalyst dose. The principal conclusions of this review are that the dye removal% is high in the early time of the adsorption operation but it reduces still it reaches equilibrium, Temperature negatively affects the dye adsorption method, there is a specific pH value for each catalyst, at which the optimum adsorption of dyes happens as well as adsorbent dose growth in general enhances catalytic activity because of the increase in total surface area and the total of active sites on catalyst surface. For optimizing the conditions for dye adsorption onto zeolite and marine algae, the factors that affect dye adsorption onto zeolite and marine algae surfaces should be known.

Keywords: Marine algae; Zeolites; textile dye removal; adsorption; catalyst dose; contact time; temperature; pH; wastewater

1. Introduction

Constantly increasing in the universal requirements for high quality as well as prospective water, increase the interests around the green treatment and reuse of wastewater [1-4]. Textile industrial wastewater is classified as the most destructive contaminations in all the industrial segments [5-7]. Several dyes that present in the wastewater are exceptionally poisonous, cancer-causing and can cause constant damage to the aquatic environments if discharged unprocessed. Dyes frequently used in different industries as cosmetics textiles and printing are toxic chemicals [8]. For dye wastewater treatment, there are a lot of technologies like biological degradation [9], coagulation [10], advanced oxidation [11], and adsorption [12], have been advanced. Among these techniques, Adsorption technique is regarded as one of the most interesting techniques since it is low-cost, easy to design, safe, leads to nontoxic substances production

*Corresponding author e-mail: ahmed.hamd@nub.edu.eg .; (Ahmed Hamd).
Receive Date: 21 August 2021, Revise Date: 03 February 2022, Accept Date: 07 February 2022
DOI: 10.21608/EJCHEM.2022.86811.4356

and effective in removal of organic pollutants and much more [13-15]. Chapman and Siebold in 1912, published The first article on dye removal by adsorption technique [16]. Because of the distinct advantages of adsorption technique, many substances had been experimented and demonstrated to be efficient in removal of dye wastes. On the other hand, 3A zeolite was used to remove Rhodamine B as this adsorbent has the ability to remove about 90% of this pollutant from industrial wastewater [17]. Also, zeolite composites showed great efficiency in removing dyes such as methyl orange and Congo red by adsorption method.[18] Macro algae have a great importance in dye removal field because they are safe, low-cost, and obtainable substances for colored wastes treatment with various extents of success [19]. The algae surface has the ability to adsorb dyes from aqueous solutions throughout maximum biosorption of Methyl Orange dye (MO) from aqueous solutions using marine alga Fucus vesiculosus biomass. The experimental results indicated that 3 g/L of F. vesiculosus biomass was capable of removing 50.27% of MO simultaneously from aqueous solution using MO (60 mg/L) at pH 7 within 60 min with agitation at 200 pm[20]. Marungrueng and Pavasant studied dye adsorption on the green macroalgae classes Caulerpa lentillifera and investigated that the sorptions of three basic dyes, Astrazon_ Blue FGRL (AB), Astrazon_ Red GTLN (AR), and methylene blue (MB) onto green macroalga Caulerpa lentillifera were higher than activated carbon when the results were compared to the sorption performance of a commercial activated carbon (CARBON). The results revealed that the alga exhibited greater sorption capacities than activated carbon for the three basic dyes investigated.[21] The purpose of this review is to assess the effectiveness of various macroalgae and natural zeolite separately and in the composite state for industrial textile dyes removal and we present definition of marine algae, classification of marine algae, definition of Zeolite , industrial wastewater and their effect on the eco system, water treatment methods, which include chemical, biological, Combinatorial method, nanotechnology-based and physical methods, uses of algae and natural zeolite in wastewater treatment and finally we discuss the factors that affect dyes adsorption onto zeolite and marine algae surfaces, such as contact time, temperature, solution pH and catalyst dose

2. Definitions and classifications

2.1. Definition of marine algae

Marine algae belong to the most exciting algae group because of their biological activities for example anti-cancer [22], anti-allergic [23], antimicrobial [24], antiviral [25], anticoagulant [26], antifungal [27], anti-fouling and antioxidant activities [28]. They produce various chemically effective metabolites in their surroundings as a defense to guard themselves from other settling beings [29]. There are numerous reports of chemical compounds derived from macro-algae that have a range of biological activities, some of which could be used in the pharmaceutical industries. Antibiotics that have the ability to inhibit bacteria, viruses, fungi and other pesticides can be produced from many seaweed. There are a lot of factors such specific algae, microorganisms, season and growing conditions that the antibiotic property depends on [30] . Grover Parul indicated that some antioxidants as β-carotene, may be suitable for cancerous conditions treatment such oral leukoplakia, which may be a precursor to cancer of mouth. [31] The progress of aquatic floral compounds as medicinal agents is still in its embryonic period due to the truth of collecting the aquatic floral samples. To separate and identify the latest natural products derived from faunal sorts, various significant efforts have been made by both medical companies and educational foundations. Marine plants have not been greatly explored to foster further research in this area. [32] Macro algae is considered as an interesting point for researchers as a lot of publications had been published all over the world as figure 1 shows according to since direct.

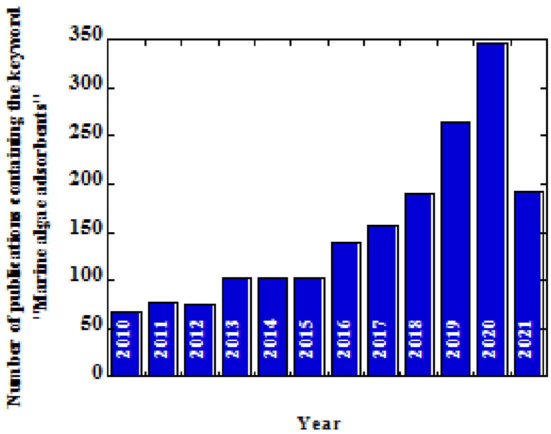

Fig. 1. Representation of the number of publications containing the keyword "Marine algae adsorbents" published from 2010 to February 2021. The data are obtained from "Science Direct".

2.2. Classification of marine algae :-

Algae, as shown in figure 2, can be broadly divided into macro-algae (macroscopic algae) and microalgae (microscopic algae). [33] Seaweed is important producers of vitamins, minerals and proteins and fatty acids, etc. [34]. Algae (macro-algae, algae) consist of about thirty thousand kinds universally. Algae are responsible for oxygen regulation in the environment, they are a source of nutrition for marine life, and are an abundant source of structurally exceptional natural products.[35]. Algae are the existing photosynthetic organisms in aquatic life. The photosynthetic algae process is similar to that of plants in a terrestrial environment. Marine plants are more operational in the change of solar biomass power. They receive nutrients directly from adjacent water through their tissues.

- Microalgae, unicellular 3-10 µm (microns) [36] comprising blue-green algae, *dinoflagelllates, Bacillariography* (diatoms), green algae and blue-green algae. They occupy the end of the nutrition chain. Despite the estimated diversity of approximately (200,000 to 800,000) species, only a portion (35) thousand is described. This is one of the most significant sources of nutrition for many organizations in the aquatic environments. The aquatic surroundings characterizes most of the algae classes[37]
- Macroalgae (seaweeds), massive kelps up to seventy meters long and growing at up to Fifty centimeters daily [38] which includes red
- (Rhodophyta), green (Chlorophyta), and brown algae (Phaeophyta) [39] In general, brown macro-algae are larger than green and red algae. The color of Rhodophyceae from time to time looks as brownish-red or purple. Algae in the marine environment are a potential renewable source and include over six thousand species.[37]

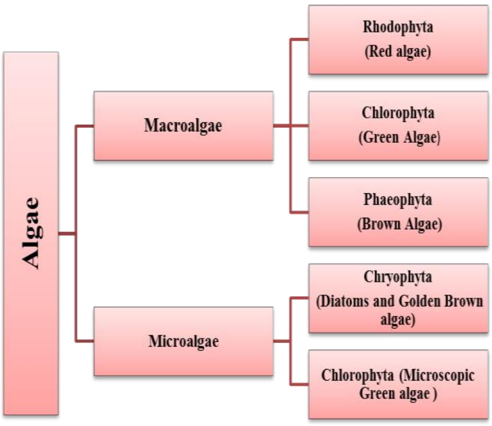

Fig .2: Schematic diagram of classification of Algae

2.3. Definition of Zeolite :-

Zeolites are biologically and economically traditional aqueous aluminosilicate substances along with particular ion substitution and absorption properties. Their effectiveness in numerous technological processes depends on their physical and chemical properties that are strongly linked to their environmental sediments. The unique tree-dimensional porous structure provides natural zeolites numerous treatment opportunities. The natural zeolites belong to the cationic exchange group as a result of the additional negative load on the surfaces of zeolite caused by the isomorphic exchange of silicon by aluminum in primary structural units. Up to now, various studies have proved their exceptional performance on wastewater treatment . However, the zeolite can be modified chemically by inorganic salts or organic surfactants that are adsorbed on the outside leading to positively charged oxyhydroxides production or surfactant micelles that lets zeolites to bind similarly anions such chromates or arsenates forming stable or less stable complexes [40]. Zeolites are commonly micro-pore raw material of silicate, varying between colorless, white and pale red probably with colors as a result of the presence of impurities and metals traces. For tribo-mechanical treatment into the patented instrument, crystal-like Clinoptilolites, have been chosen, generally as a result of their features of selectivity, capacity of ion substitution, and absorption ability. Through chemical and toxicological studies achieved by scientists, it has been demonstrated that Clinoptilolites are generally nontoxic [41]. Natural zeolite is considered as an natural resin and cost-effective because it discharges potassium, calcium, sodium along with magnesium ions toward the surroundings, these ions aren't poisonous [42, 43]. There are various types of zeolite have been identified, however some of zeolite mineral deposits form the main segment of volcano deposits only: analcime, chabazite , heulandite, erionite, clinoptilolite, ferrierite, mordenite, laumontite and phillipsite. The structure of these minerals is various, but all of them have big open channels in the crystal constitution that afford a large empty space for cations adsorption and exchange .[44] Zeolite is considered as an interesting point for researchers as a lot of publications had been published all over the world as figure 3 shows according to since direct.

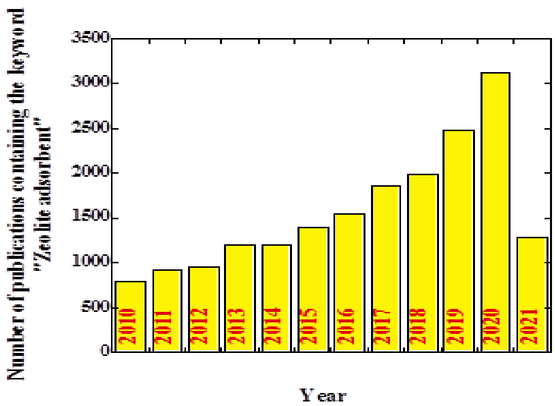

Fig. 3. Representation of the number of publications containing the keyword "Zeolite adsorbent" published from 2010 to February 2021. The data are obtained from "Science Direct".

3. Industrial wastewater and their effect on the eco system

In table 1 a lot of pollutants produced during different textile dyes Processes and have harmful effect on the eco system . The treatment of wastewater involves the rupture of complex biological compounds in wastewater in simpler stable and non-nuisance compounds, or physico-chemically and / or using microorganisms (biological treatment). The negative impact on the environment to allow the evacuation of untreated wastewater in groundwater or surface water and or land are as follows.:

1. The decomposition of organic materials contained in wastewater may result in the production of large amounts of smelly gases.

2. Wastewater containing a great quantity of organic substance, if released in the marine environment, the dissolved oxygen will be consumed to satisfy the demand for biochemical oxygen (BOD) of wastewater and thus exhausting the dissolved oxygen from the flow, thus causing killing fish and other adverse effects.

3. Sewage can also contain nutrients, which can accelerate the growth of aquatic plants and algae flowers, resulting in lakes and streams eutrophication

4. The wastewater generally contains many pathogenic microorganisms or causes toxic disease and compounds, which live in the individual intestinal tract or may be existing in certain industrial waste. These can contaminate the earth or the body of water, where such wastewater is eliminated. For the reasons mentioned above treatment and removal of sewage, not only desirable but also necessary.[45]The textile dyes, together with several industrial contaminants, are extremely toxic and highly carcinogenic [46] because they are associated with the degradation of environment and several diseases in animals and persons [47] The tendency to rebel in aerobic ecosystems, particularly in conventional treatment stations, is responsible for collecting vital dyes in sediments and soils and carrying them to supply systems of public water [48]. Even though the environmental majority recalcitrance, they may be partially destroyed or converted in the presence of anoxic sediments, as happens in the azo compounds reduction causing hazardous aromatic amines [49]. A further probability is to combine dyes together with intermediate synthetic compounds or the products of their degradation to yield other cancerous and mutagenic materials [48].

1. Water treatment technologies

Treatment of wastewater generally includes neutralization and elimination of dangerous chemicals and colors. Several methods are accessible with competitive Pros and cons Sewage is treated at numerous stages[50]. In the initial stage, after the removal of very rough solids, adjust capable solids involving organic and inorganic substances are eliminated. Most of the Dissolved Solids (DS), Suspended Solids (SS), greases and oils are separated from sewage.

Table 1. Pollutants in Wastewater at Different Textile dyes Processes:-[51, 52]

processes	**pollutants in wastewater**	**quality parameters to assess environmental impacts**
Sizing	Starch, waxes, carboxymethyl cellulose, polyvinyl alcohol (PVA), wetting agents	High Biological Oxygen Demand (BOD), Chemical Oxygen Demand (COD), high turbidity, low Dissolved Oxygen (DO), softener, oils, fats
Scouring and bleaching	Sodium hypochlorite, Cl_2, NaOH, H_2O_2, acids, surfactant, Na2SiO3, sodium phosphate, microfibers	High temperature, high alkalinity, high SS, pectins, proteins, oils, silicates, high COD
Dyeing	Dyestuffs, urea, reducing agents, oxidizing agents, acetic acid, detergents, wetting agents	High temperature, strong color, high BOD, high DS, low SS, low heavy metals, high salinity, high electric conductivity, low DO, high turbidity, high Total Volatile Solids (TVS)
Mercerizing	NaOH, cotton wax	High pH, high BOD, high DS
Desizing	Starch, carboxymethyl cellulose, PVA, fats, waxes, pectins, enzymes, hemicellulose	High BOD, COD, Suspended Solids (SS), Dissolved Solids (DS), high turbidity, low DO, acidic pH
Printing	Urea, starch, gums, oils and greases, binders, fixers, acids, thickeners, cross-linkers, reducing agents, alkali	Highly colored, high BOD, oily appearance, high SS, slightly alkaline, high turbidity

In the secondary stage, soluble and non-soluble contaminants are eliminated via biochemical methods such as aerobic and anaerobic. Lastly, The third and sophisticated phase is very object oriented and used to eliminate dyes and other poisonous substances.[53-55]. The third and improved technology is sophisticated, and several processes of this type are represented in Figure 4. Wastewater treatment classification depends on interaction and mechanism type for example physical, biological and chemical. The subclasses below the wide classification have been reported in different ways in the works.[51, 56-59]. The irregularity is found because of several reasons; For example, only one class can represent numerous techniques and several classes are combined to serve only one class. It can classify in three processes depending on contaminants nature: chemical, physical and biological [60-62]. The advantages and disadvantages of the different techniques of wastewater treatment as shown in table 2

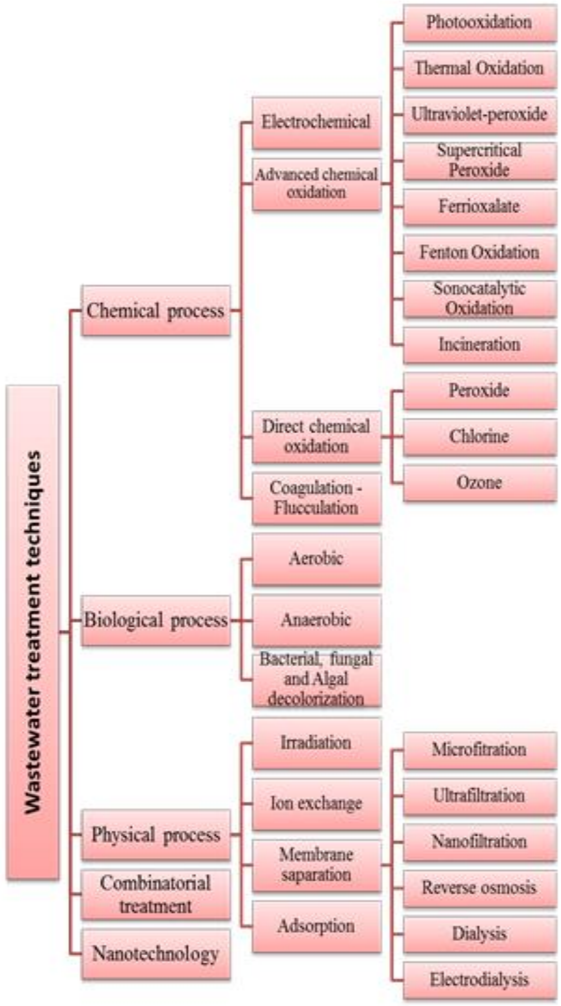

Figure 4. Schematic diagram of classification of wastewater treatment techniques

1.1. Chemical process

The chemical technique includes the use of chemical substances performing chemical reactions for neutralization, oxidation and wastewater disinfection [63]. The major classes of the chemical process are chemical oxidation as well as advanced oxidation methods. They may be used separately or in combination with each other, often called mixed advanced oxidation methods Oxidation methods involve ozone, Fenton, sono catalytic and photocatalytic oxidations, in which hydroxyl radicals that have very high oxidation potentials are created.[63, 64] These radicals attract and oxidize a majority of organic and inorganic compounds in textile effluents at a very high rate. Additional potential advanced chemical process is the photo degradation method that the phenomenal results by eliminating wastewater contaminants.[65, 66]. Researchers showed the complete removal of textile wastewater solution colors applying the photocatalytic oxidation process.[67-69] Lately, besides 100 %, removal of the wastewater solution color on laboratory such as Basic Red 18 by zinc oxide impregnated on fungi, [69] complete elimination of colorants from actual industrial wastewater has been determined employing the S_2O_8 $2-/Fe^{2+}$/UV method.[68] However, integration of further methods as sonication is able to enhance degradation performing since reaction time is long as well as the consumption rate of catalyst is high.[70, 71] In addition, the application of ultrasonic technology is mainly pronounced to expand the improvement of adsorption capacity by improving the rate of mass transfer on the adsorbent used radically.[72] Coagulation and flocculation combination is widely used for the treatment of wastewater to eliminate most colloidal, suspended and dissolved solids. Managing by-products such as green gases and sludge of different chemical processes is always difficult and a cause of an great ecological trouble. In addition, most of these methods are high in intensive energy and subjected to the type of contaminant and, therefore, are not economically viable.[58, 73] The interaction of the dye surfactant, a separate chemical mechanism beyond conventional chemical processes, could be a promising method for wastewater treatment. A suitable surfactant is used for removing desired dye molecules from wastewater. When the surfactant molecules have been added to wastewater forming a three-dimensional structure in the system and absorb the dye molecules in their interiors or adsorb on the surface by dye surfactant interactions. These bodies of dye surfactants can be simply removed by several physical processes and thus the wastewater is discolored. This method is very effective and capable of eliminating 100% wastewater dyes with a generation of minimal sludge. The dyes and surfactants can be completely recovered and reused. [74, 75]. This method is very efficient for ionic and nonionic dyes. In particular, it is an effective method for removing water-soluble dyes, such as ionic dyes that are difficult to eliminate effluent. In addition, the treatment of wastewater containing mixed dyes is tedious after other processes, but can be efficiently achieved by interaction of dye surfactant. [74-77] In addition, the surfactant added in textile methods can help wastewater treatment with the surfactant to be used in the treatment bath and save the cost of the method. Thus, following an interaction of surfactants with high effectiveness, profitability and sustainability may be a deserving method in a pragmatic way for the treatment of wastewater.

Table 2: Advantages and disadvantages of wastewater treatment processes

Process		**Advantages**	**Disadvantages**	**References**
Chemical process	Coagulation –flocculation	▪ Elimination of insoluble dyes and heavy metals ▪ is in extensive use for pre-, main or posttreatment, and full decolourization is possible by this process	▪ Production of voluminous sludge ▪ is not always effective ▪ there are problems associated with sludge disposal	[63-65]
	Advanced oxidation processes	▪ Are possibly the best technologies to totally eliminate organic carbons in wastewater	▪ only effective in wastewater with very low concentrations of organic dyes. Thus, significant dilution is necessary as a facility requirement. ▪ Too expensive and complex at the present level of their development	[63,66-69]
	Electrochemical processes	▪ Capacity to adapt to different pollution loads and volumes. ▪ Low ferrous oxide sludge, (Chemical Oxygen Demand) COD reduction, colour removal ▪ Breakdown compounds are nonhazardous	▪ High Energy consumption of 1 – 2 kWh/ m3. ▪ Ferrous oxide sludge ▪ High cost of electricity	[70,71]
	Ozone treatment	▪ Good decolourisation and (Chemical Oxygen Demand) COD reduction ▪ Applied in gaseous state; no alteration of volume	▪ Cost of treatment ▪ Short half life	[72,73]
	Fenton's reagent	▪ Effective decolourisation of both soluble and insoluble dye	▪ Sludge generation	[74]
Biological Treatment	Aerobic	▪ Highly efficient treatment method ▪ Requires little land area ▪ Applicable to small communities for local-scale treatment and to big cities for regional-scale treatment ▪ Minimal land requirements; can be used for household-scale treatment ▪ Relatively low cost and easy to operate ▪ Low capital cost ▪ Low operation and maintenance costs ▪ Low technical manpower requirement	▪ May produce undesirable odors ▪ Requires a large area of land ▪ Requires mechanical devices ▪ Requires technically skilled manpower for operation and maintenance ▪ High cost ▪ Requires little land area Requires sludge disposal area (sludge is usually land-spread)	
	Anaerobic	▪ Biological mediated process ▪ Recovery of non-conventional energy ▪ Methane recovery ▪ Capable of being simulated for treating ▪ wastes emanating from municipal, agricultural, and industrial activities ▪ High reduction of biochemical oxygen demand ▪ Low sludge production ▪ No electrical energy is required	▪ Energy required ▪ Sludge disposal is minimal ▪ Long start-up phase ▪ Low reduction of pathogens and nutrients ▪ Effluent and sludge require further treatment	

		▪ Simple to operate		
Physical Process	Adsorption on activated Carbon	▪ Good removal of a variety of dyes	▪ Costs for polymers and chemicals ▪ Very expensive to operate	[74-76]
	Membrane filtration	▪ Removes all dye types	▪ Concentrated sludge production	
	Ultrafiltration/microfiltration	▪ Low pressure membranes improves turbidity of effluent	▪ Produces water of lower quality than RO/NF. Cannot be used alone without biological treatment. Prone to fouling of membranes	
	Nanofiltration	▪ Separation of low molecular organics and divalent ions.	▪ Energy intensive due to high pressure requirements but requires less power than for RO membranes	[77]
	Reverse Osmosis (RO)	▪ Removal of all mineral salts. ▪ Water of high purity is produced	▪ Prone to poisoning of membranes by cationic surfactants if used without biological processes. ▪ Energy intensive due to high pressure requirements	[78-80]
	Ion exchange	▪ Regeneration; no adsorbent loss	▪ Not effective for all dyes	[74-81]
	Irradiation	▪ Effective oxidation at lab scale	▪ Requires a lot of dissolved O2	[74]

1.1.1. Advanced Oxidation Process

An advanced oxidation method can be used to eliminate wastewater dyes for production of a very reactive radical that can react a broad variation of compounds difficult to decompose. This method involves chlorination, photocatalytic oxidation and bleaching [78]

1.1.2. Electrochemical process

The process of electrochemical destruction is a relatively new technique in which relatively little or no chemical consumption is required. Apart from this, the process eliminates the sludge assembly. In the case of an electrochemical method, organic pollutants are adsorbed on the surface of the anode followed by oxidation. The cathodic reduction of oxygen with hydrogen peroxide can also oxidize many azo dyes efficiently. Another tactic explains Fe catalyst use for hydrogen peroxide electro generation leading to enhancement of dyes degradability through numerous folds. But this method is accompanied by certain troubles for instance the requirement of electrolytic device, expensive electricity, and reduced dyes decolorization as flow rate is higher [79-81]

1.1.3. Fenton's process

The oxidation process depends on Fenton's reagent (a combination of H_2O_2 and iron salt) which was used to treat organic matter and inorganic materials. The method is based on the configuration of interactive oxidation types capable of disintegrating pollutants effectively on wastewaters [82]. It is recognized that hydroxyl and ferrylic complexes happen in Fenton reaction depends on the mechanism and conditions of use of which one is predominant [83]. The oxidation system can be used efficiently for the destruction of non-biodegradable contaminated wastewater [84]. Fenton's oxidation process can stain a great variety of dyes and relative to ozonization. The method is comparatively low-cost and, in general, leads to a reduction in higher chemical oxygen demand [85]. Fenton oxidation is limited to that of the textile process; When wastewater is generally high at pH, while the Fenton process requires low pH. At the higher pH, deposits produce large amounts of iron salts of waste liqueurs and the process loses its effectiveness. $\mathbf{Fe + H_2O_2 + hv \rightarrow OH^{\bullet} + OH^{\bullet}}$

$$\mathbf{organic\ polutent^{+\bullet} + OH^{\bullet} \rightarrow Degradaed\ products}$$

Fenton reagent (Fe^{2+} & H_2O_2) affords an appropriate chemical ways of manipulating soluble and insoluble dyes in waste water, that are strong to biological treatment. Fenton's reaction was initially investigated by H.J. Fenton on 1894. It has been found that the oxidative potential of hydrogen peroxide is improved when the iron (Fe) is used as a catalyst only in acidic media. A disadvantage of this system is the unnecessary creation of sludge, which generates challenging elimination. In addition, the method demands a long time working and can't target vat and dispersion dye [81]

1.1.4. Photochemical process

In this treatment method, light (UV, visible, or solar radiation) is used with Fenton's process or without Fenton's reagent. In this process, light activates the generation of hydroxyl radicals to mineralize dye molecules to CO_2 and water [86]. The disadvantages include formation of colorless byproducts, which may be more noxious than the original dye. However, numerous research works undergoing in this arena might end up with a solution.

$$\mathbf{H_2O + hv \rightarrow H^{+} + OH^{\bullet}}$$

$$\mathbf{organic\ polutent^{+\bullet} + OH^{\bullet} \rightarrow Degradaed\ products}$$

1.1.5. Oxidation:

In these methods, chemicals such as sodium hypochlorite, hydrogen peroxide, (O_3 & H_2O_2), K_2FeO_4, or $KMnO_4$, and so on are used to transport the degradation method. The synergistic action of ozone and hydrogen peroxide causes faster generation of hydroxyl radicals, responsible for the oxidation of contaminants in water [87]. The characteristic reaction is indicated as follows:

$$2O_3 + H_2O_2 \longrightarrow 2{\bullet}\,OH + 3O_2$$

The oxidation of textile waste with these chemicals happens at a smaller time, but the process is expensive and depends generally on pH medium. The chlorination of dye wastewater by effective chlorine is infrequently used because of its harmful effects on its flow in the waterways. [88]

1.1.6. Ozonation

Ozone is a strong oxidative agent that ozone has a high oxidation potential (2.07) compared to hydrogen peroxide (1.78) and chlorine (1.36). A rapid reaction happens as well as no generation of sludge happens. a main gap of this method is the rapider half-life (twenty minutes) of O_3 and therefore requires a constant effective rate of incessant stream. Additionally, the method is sensitive to temperature, salts presence and pH. Ozone generates oxidative radicals in waste water, resulting in the chemical degradation of contaminants. The Typical mechanism of the reaction that happens throughout ozonation process is described as follow [89]

$$3O_3 + O\bar{H} + H^{+} \longrightarrow 2\,\dot{}\,O\bar{H} + 4O_2$$

Biological Method

The biological process is mainly preferred to eliminate biodegradable elements in waste water. Microorganisms degrade organic pollutants and reduce demand for biochemical oxygen and suspended substances, [58] and the effectiveness of elimination is dictated by the charge ratios of the organic, microorganisms and dyes.[59] Anaerobic and Aerobic are the main categories of the biological process.[57] According to demand oxygen types and amount in addition to wastewater nature, a mixture of aerobic and anaerobic methods can be operated. The advantages of biological methods are ecological, economic sludge, minimized sludge and a generation of non-hazardous metabolites.[64, 94] Nevertheless, the complexity in creating a pleasant environment, slow processes and poor discoloration limits the commercial acceptance of biological methods. In addition, biological methods sometimes do not show the desired effectiveness due to the non-biodegradability of synthetic dyes.[58]. In general, Polymers as well as dyes are hard to biodegrade and several materials are inappropriate for traditional biological treatments. Particularly, For textiles dyes, more importance is provided on methods of biological treatment with respect to physical and chemical processes. Generally, the techniques of biodegradation are pragmatic in the treatment of industrial effluents many microorganisms such as bacteria, yeasts, algae, and fungi may accumulate and damage different contaminants [61], and all biological methods involve continuous sewage. Biological treatment needs a great land space in addition is limited via sensitivity towards the daylight change and chemicals toxicity and less flexibility during design and process. Biological treatment is incapable of gaining suitable color elimination by recent traditional biodegradation methods.

1.1.7. Removal of dyes by Bacteria

The power of bacteria to absorb dyes such as azo has been examined by many researchers [62]. In aerobic environments, azo dyes are not instantly absorbed, although the capacity of the bacteria with specialized reducing enzymes to be degraded aerobically certain azo compounds [63]. On the other hand, many bacteria are anaerobic that reduce AZO dyes by non-specific and soluble activity, cytoplasmic reductase. Anaerobic reduction of degraded azo dyes can convert to aromatic amines [64], which can be toxic, the complete degradation of azo dyes or AZO compounds requires aerobic biodegradation of materials produced [66]. In phthalocyanine colors, two-sided reduction and decolorization are existing in anaerobic environments [67]. Mutagenic, and probably cancer-causing for creatures [65].

1.1.8. Removal of dyes by Fungi

The highly examined fungi about dyes degradation are ligninolytic fungi which create enzymes as laccase, manganese peroxidase, and lignin peroxidase [95]. There are several works on the fungi capability of oxidizing phenolic, nonphenolic, soluble, and insoluble colorants [96] Types of *Pleurotus ostreatus*, *Neurospora crassa*, *Schizophyllum* and *Sclerotium rolfsii*, existed to grow up to quarter decolorization degree of specific colorants such as triaryl methane, indigoide and anthraquinone applying enzymatic preparations [97]. On the other hand, manganese peroxidase has been reported as the major enzyme included in removal of color by *Phanerochaete Chrysosporium* [98]

besides lignin peroxidase for *Bjerkandera adusta* [99] The researchers also mentioned that some colored mushrooms are able to remove color pigments effectively [100]

1.1.9. Removal of dyes by Algae

There are a few algaes like *Chlorella, Oscillateria [101] and Spirogyra* [102] are indicated for dyes degradation. Additionally, Jinqi and Houtian [101] reported that specific azo compounds examined can be used as nitrogen and carbon sources. This can mean that algae play an significant role in removing aromatic amines and azo dyes in stabilization ponds [78].

1.2. Combinatorial treatment method

Not any of the treatment processes discussed previously is devoid of disadvantages. These methods do not fail either of a variety of substrate and force sufficiently to eliminate the poly aromatic dye in waste water. A possible solution to resolve this problem is the successive application of a method combination. These treatment processes must be crucially designated so that the drawback of one method may be overcome via the other [103]. Many combinatorial processing methods are in the investigation and test phase in research laboratory. The high sensitivity, the lack of integrity and the slow of the biological method mainly appreciate the usage of physicochemical treatment systems for combinatorial determinations. Another literature recommends adsorbent coalescence characteristic, physical method, by a bio catalyzer , biological enzyme method, being as a very commercial and need easy handling [104]

1.3. Nanotechnology-based treatment techniques

Nanoscience has attracted the growing attention from researchers around the world to investigate the potential of nanomaterials in the treatment of wastewater. Several inorganic nanotic materials, such as metal, metal oxide, metal sulfide materials, etc., find an application towards the elimination of dye pollutants. The following section provides a brief insight of these materials and their applicability for the treatment of wastewater. Control factors that cause upper removal efficiency are a high surface area, crystalline, surface load, band space and specific affinity. The key advantage of the treatment techniques based on nanotechnology lies in several cycles of catalyst reuse, resulting in the cost of the method. Furthermore, the use of Nano catalyst gives a minimal ecological effect, low solubility and a generation of pollutants to zero or few secondary contaminants.[105-107]

1.4. Physical process

The Physical treatments of waste water are possessed via van der Waals forces, gravity, along with electromotive forces, or via physical blocks, physical state converting.[63, 64] The conventional physical treatment processes are filtration, ion exchange, an irradiation and adsorption method.[55, 56, 63, 108, 109]. However, the utmost of physical systems are appeared to indicate low efficiency of dye elimination and generate a large quantity of sewage, the adsorption method is highly preferred due to its exceptional dye removal capability.[72] On the other hand, the variety of adsorbents and its concerning factors, reconstitution, and/or removal of adsorbents and a great quantity of sludge creation have limited broad use of the adsorption method.[58] Fractionation of dye together with further smaller chemical classes may be captured efficiently with extremely great effectiveness up to 99% via membrane separation methods such as reverse osmosis, nanofiltration, ultrafiltration and microfiltration. For example, the treatment of integrated ultrafiltration diafiltration membranes can recover more than 97% of the dye and attain more than 99% desalination.[110, 111] Nonetheless, due to the requirement of high-pressure application, membrane separation processes are not cost-efficient.

1.4.1. Ion-exchange

Ion replace is a reversible water treatment process containing ion-exchange resin that swaps away one or more undesirable contaminants in exchange with another non-objectionable or less objectionable substance. Therefore, the resin is regenerated via a continuous backwashing method with an intense solution of exchange ions to eliminate collected ions followed with dousing the flushing solution from the resin. Certain modified natural substances, for instance, organofunctionalized covered silicate and cationic-polymer/bentonite complex, exposed remarkable results [112]. The need for counter-current washing, rinsing and rinsing during the renovation of the ion replacement media limits the practice of the exchange of ions for the treatment of wastewater. Although loaded dye substances can be eliminated by this method, the process is not exceptionally compatible with dye elimination. [113].

1.4.2. Irradiation

Among the several systems for treatment of wastewater, the irradiation technique with ultrasound or microwave is well-known. Ultrasound frequencies of a certain power are emitted into the water by precise transducers. Transient cavitations, improved by the successive scarcity and density of bulk water, when collapse generates an elevated temperature and local pressure peaks. This separates water molecules into hydroxyl radicals and hydrogen atoms. The formed radicals, pressure and temperature are able to damaging dye contaminants. High frequency ultrasound can considerably degenerate the color of colorants. Addition of Titanium dioxide or zero-valent copper, Carbon tetrachloride improved low frequency ultrasound performance. In this scenario, a enormous quantity of liquefied oxygen is needed, which restricts its application on a great scale [114] Lately, microwave irradiation has drawn a lot of attention to sewage treatment applications. Several microwave absorbing materials, as well as the high surface area and a wide range of pore size distribution, have been progressed to improve the degradation of organic contaminants in microwaves [115] When this substance is used, heated spots are created upon the surface causing selective warming, leading to degradation. These processes necessitate elevated energy as well as can generate poisonous and cancer-causing aromatics through pyrolysis.

1.4.3. Membrane filtration:

Membranes recommend amazing probabilities for dyes separation and coloring auxiliaries from water. Numerous materials like ceramic films prepared from clay and alumina, and nano filtration polyamide-based composite films have revealed decent decolorizing properties, especially when removed after coagulation and flocculation [116] The pros of membrane filtration are a rapid, low space method needing space as well as may be reprocessed. The chief drawback of the filtration process of the film is its brief lifetime, because of contamination, which extremely improves the method effectiveness. [117] The film filtration can be various types like ultrafiltration of reverse osmosis and nanofiltration. In the old method, dyes pollutants are allowed by an enormously decorated film and consequently, needs elevated pressure for separation pollutants from treated water. In addition, the membrane is often clogged with dye particles. As a general rule, reverse osmosis is a pressure controlled system where the liquid is allowed to pass from the low solute concentration to a highly thinly solute concentration by an extremely thin partial permeable film with a maintenance degree of 90% or more. Effectively, Reverse osmosis isolates pollutants on single side of the film and treated water on the further side. Remember that the higher the solute concentration, the greater the osmotic pressure and therefore greater energy is mandatory for the separation method. This limits its applicability because there is a constant high pressure requirement, which makes the process expensive [118].

1.4.3.1. Ultra Filtration

Ultrafiltration permits the elimination of particles as well as macromolecules, however the removal of contaminants like dyes isn’t completed [119] the quality of treated wastewater does not allow the reprocess of sensitive methods like textile dyeing, except from 31% to76% in the effective cases. Ultra filtration doesn’t have the ability to be used as a pre-treatment for reverse osmosis [120]or with a biological reactor [121]

1.4.3.2. Microfiltration

Microfiltration is appropriate for the treatment of colorants having pigment dyes [122] in addition to successive soaking baths. Compounds consumed in the coloring bath, which is not clean with using microfiltration process, will stay in the bath. Additionally, the process of microfiltration may be used in pretreatment of nanofiltration or else in reverse osmosis [123]

1.4.3.3. Nano filtration

Nano filtration has been used for removing dyes from wastewater. The step of adsorption precedes nanofiltration since this system decreases the concentration polarization throughout the filtration method, which rises process production [124] Dangerous results of elevated concentrations of colorants as well as salts in dye house drainages have been informed [125-127]. In most studies published on dye Sewage, the concentration of mineral salts is not more than 20 g/l and the concentration of dye does not surpass 1.5 g/l [128]. Generally, wastewater with single dye can be re-formed [129], besides, the studied volume studied is low too. [130] An important problem is the accumulation of dissolved solids, nanofiltration is considered as one of the rare applications which can be used for treating solutions with very concentrated as well as complex solutions which removes the treated discharge drainages in water flows .

1.4.3.4. Reverse Osmosis

In reverse osmosis, films have a retention rate of 90% or more types of ionic compounds as well as generate great quality permeate [118]. Degradation besides removal of chemical additives inside colorant sewage may be processed in only one step reverse osmosis. Reverse osmosis makes it probable to hydrolyze all mineral salts and reactive colorants and chemical aids. It should be observed that the higher the concentration of dissolve salt, the greater the osmotic pressure; Therefore, the more energy needed for the separation method is great.

1.4.3.5. Electro-Dialysis

The electro dialysis (Ed) is a membrane separation method used to the separate the anions and cations using double membranes loaded with anode and cathode. It is a very applicable and appropriate method for inorganic as well as harmful contaminants management [131] The membranes are generally ion-exchange resins that move ions in a selective way. They are produced from a polymer of substances, like polyethylene or styrene, combining with stationary and movable charged groups [132, 133]. Throughout this method, positive ions transfer to a negative cathode plate as soon as the electrical current passes on an aquatic solution of metal [134]. Furthermore, ED needs a greater frequency of pretreatment before the method. The operative effectiveness of ED depends on several features, like membrane quality, current intensity, pH, structure of ED cell, concentration of H_2O ions, as well as current rate [135] ED deals with the harmful contaminants which are present in the shape of metals, solids plus other constructs of H_2O in the industrial textile wastewater. Additionally, it is used to recuperate certain beneficial metals like copper and chromium. A observable disadvantage of this method is the erosion and polluting of film influenced via solid elements or colloids as well as biomass which decrease the transportation of ion [136]

1.4.4. Adsorption:

Physical adsorption confirms weak intermediate bonds between the adsorbent and the adsorbate. Feeble physical forces like hydrogen bonding, Van der Waals' interactions, and dipole-dipole interactions commonly take part. In general, physical adsorption is definitely reversible, but irreversible chemisorption happens when great bindings present between the adsorbate and the adsorbent through electron replacement. Most adsorbents are porous substances with great surface area and optimal pore diameter. Amongst several methods of colorant removal, adsorption method can be used for dye removal from industrial textile wastewater significantly [137]. Additionally, This method is affected with numerous factors like adsorbent surface, interaction of dye and adsorbent, temperature, interaction time, particle size and pH. These methods with the properly designed technique can eliminate colorants existing in wastewater. In this process, ions or molecules existing in only one phase are inclined to gather and collect on the surface of another phase. Physical adsorption happens when feeble transitional bonds exist among adsorbate and adsorbent. Instances of these bindings are Van der Walls interactions, dipole-dipole interactions, and hydrogen bonding. In most cases, physical adsorption is definitely reversible. Chemical adsorption occurs when powerful bindings are existing between the adsorbate and the adsorbent via replacement of electrons. These bonds can be covalent and ionic. Chemisorption is irreversible in several cases. Suzuki [138] examined the adsorption role in marine ecosystem methods and similarly estimated the improvement of latest adsorbents to enhance the treatment and techniques. Several adsorbents are extremely spongy substances. Because the pores of adsorbent are usually very little, the inner surface is in the order size larger than the outside area. Among several methods of colorant elimination, adsorption method provides the greatest significant outcomes since it is able to be used for eliminating all types of dyes [138]. Because of great efficiency of adsorption technique in the removal of pollutants that are not readily biodegradable, production of high quality water as well as profitable, these adsorption methods have become very general in latest days. Color removal is a result of two mechanisms - adsorption and ion replacement and is influenced by several factors involving interaction between dye and adsorbent, adsorbent surface, particle size, temperature, pH and contact time. If the adsorption method is appropriately designed, it will yield a high quality decolorized waste water.

2. Mechanisms of biosorption:

As demonstrated in figure 5 system of color biosorption by algal biomass macroalgae walls are basically comprised of alginate, fucan, proteins and afterward the main functional groups should be carboxyl, sulfate and amine[139, 140] In acidic arrangements the protonation of amine capacities permits the electrostatic attraction of color particles that are negatively charged (sulfonic groups). The mechanism of electrostatic attraction between anionic colorants and cationic surface of the biomass in acidic preparations may clarify the higher effectiveness of color bio-sorption at pH under 4. The role of Proton (H^+) is to link between the algal cell wall and the colorant particles [140]. At the point when the pH increments (above pH 6) the amine groups are deprotonated (diminishing the tendency of the sorbent for the colorant) while other useful groups, for example, carboxylic acids are negatively charged expanding the electrostatic aversion between anionic colorant and the anionic reactive groups. [140] This outcome is steady with concentrates on the adsorption of AB1 onto chitosan, three diverse macroalgae and unburned carbon [141-143]. Therefore, adjustment of zeolite by algal biomass improved the adsorption capacity of zeolite to enormous degree .[144]

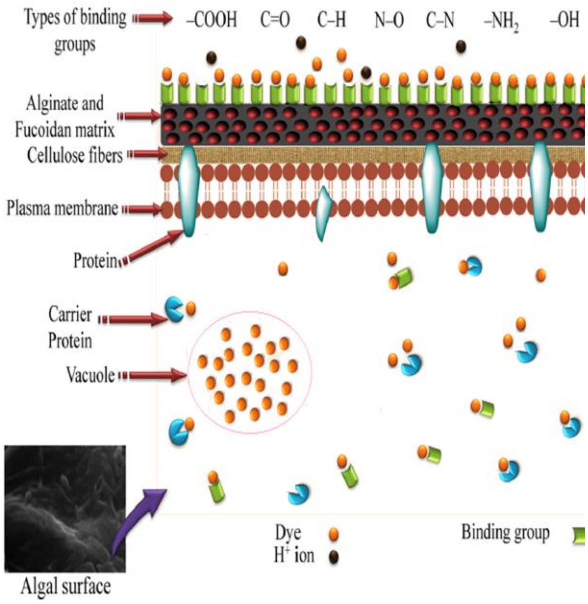

Figure 5. Biosorption mechanism of dye by algal biomass.[20]

3. Use of algae in wastewater treatment

As algae has the ability to collect vegetable nutrients, heavy metals, pesticides, inorganic noxious materials, and radioactive substances in their cells or bodies they have become significant creatures to treat industrial wastewater naturally [145-148]. **Organic** wastewater treatment systems with microalgae have become principally significant fifty years in addition to largely recognized that algal sewage treatment processes efficient as traditional techniques of treatment. These particular features have made algae sewage treatment processes a considerable cheap alternative to the high-priced complicated treatment techniques particularly for wastewater purification. Additionally, algae collected from treated tanks are usually used as phosphorus along with nitrogen supplement for farming aims and may be exposed to fermentation to get power from methane. Algae are able to exceptionally collect toxic materials such as selenium, zinc and arsenic into their cells consequently remove these materials from aquatic ecosystems. Numerous algae are capable of absorbing as well as collecting numerous radioactive metals inside their cells, even at greater concentrations in water [149]. Mackenthun indicated that Spirogyra alge can collect radio phosphorus by a factor 85×10^3 times that of water [150]. It has been observed that algae could remove dye substances from Sewage efficiently. Mohan et al [151] studied that the alga Spirogyra was noticed to eliminate the azo dye reactive yellow 22 professionally. In the same way, Daneshvar et al [152] presented the elimination of malachite green via Cosmarium sp. In a further investigation by Sarwa et al[153] the microalga Acutodesmus obliquus was observed to eliminate acid red 66 azo dye from industrial waste water. In the same way, Chlorella vulgaris was discovered to eliminate Congo red with dye removal rate reached up to 83% at an initial concentration of 5 mg/L as well as 53% at a concentration of 25 mg/L [154]. Additionally , Tetraselmis sp. and Nannochloropsi sp., were obtained to be utilized in dye elimination from industrial waste waters [155].

Agreed with all of these algae's ability for wastewater decontamination, it should be highlighted that algae technique in wastewater treatment processes have to become more significant in the following days.

Maximum sorption capacity (qm) got from the sorption of methylene blue (MB) on the sutface of green macroalga Caulerpa lentillifera was more than 400 mg/g which was comparatively higher than activated carbon . El-Sheekh et al examined that green algae as well as cyanobacteria were significant hotspots for dye removal. Chlorococcum sp., Scenedesmus obliquus, Oscillatoria sp as well as Chlorella vulgaris for degradation along with elimination of certain azo dyes such as Reactive Orange 122 (Orange 2RL) besides Reactive Red 194 (Reactive Red M-2BF) their outcomes presented that the maximum color removal was spotted at 20 ppm Reactive Orange 122 with Oscillatoria sp. mixed with S. obliquus 98.54 percentage. 20 ppm Reactive Red 194 was eliminated through Oscillatoria sp. mixed with S. obliquus (97.58%) after incubation for seven days. [156]. Additionally, It has been noticed that macroalgae have a colorant adsorption capacity on surface of green marine algae. For instance, Ulva lactuca was used as the natural resource for the formulation by impregnating Zinc chloride as well as the nano-activated carbon (IUAC) had exhibited a favorable adsorption capacity for methylene blue (MB) removal, a low-price nanostructured activated carbon (IUAC). It has a great surface area and the maximum adsorption capacity (Qm) of MB was 344.83 mg/g at room temperature. Therefore, it was confirmed that Ulva lactuca may be used in the same way as a favourable natural resource for the creation of efficient activated carbon including a particular great surface area (1486 m2 / g).[157]

4. Using zeolite in treatment of wastewater

The use of natural zeolites in sewage treatment is one of the fundamental and the most side areas of their treatment. The existence of Congo red dye in wastewater is hazardous environmental problem as well as its elimination by using natural zeolites has been considerably investigated besides other methods, such as membrane filtration, chemical precipitation, adsorption, flotation ion exchange, coagulation-flocculation and electrochemical processes [158, 159]. Current investigations of natural zeolites as adsorbents in treatment of waters, their Characteristics, and natural zeolite modification have been a subject of several studies. All over the world, Numerous natural zeolites have exhibited effective ion exchange ability for cations such as ammonium. Modification zeolites can be attained by numerous methods such as surfactant functionalization, acid treatment, and ion exchange. The modified zeolites can show good adsorption capacity for organic substance along with anions [160].

4.1. Natural zeolites in water treatment

Over the last ten years, numerous outcome results have exhibited that natural zeolites have practical use, which is recognized through a large number of patents, particularly for the two raw materials of natural zeolite such as mordenite as well as clinoptilolite. The total of patents is important for two types of zeolite that gives an noticeable sign that the consideration of investigators in natural zeolites is extremely inspired by the industrial segment covering the use in homes or industrial significant techniques and treatments.[161]

4.2. Natural zeolites in drinking water treatment

The natural zeolite is extremely used for wastewater treatment such as drinking water surface water, grey water and underground water. The first process for the treatment of wastewaters involved using of natural zeolite. Furthermore, natural zeolite has the ability to be used for removing heavy metals such as Cd, Cu, Mn, Zn, Cr, Pb, and Fe.[30-32]. Additionally, the natural zeolite is used with other technologies (ion exchange, membrane filtration, flotation, and coagulation). Natural zeolite has effective cation exchange abilities [162]. For treatment of surface water, the use of zeolite exhibits the greatest outcomes for eliminating acid along with ammonia while keeping the pH near to natural water. Natural zeolite is used also for removal of Mn as well as Fe. [163]. Furthermore, in drinking water and gray water treatment, natural zeolite is used as mineral absorbent. Arsenic is exceptionally poisonous in drinking water and is removed by using natural zeolite. It eliminates Cu from drinking water. Groundwater includes elevated levels of F^- up to 30 mg / L in Africa, Asia and the United States. As for gray water, it is attained from cleaning and bath drains. It should be observed that a huge quantity of ammonium is existing in gray water. Natural zeolite using allows elimination of ammonium from graywater [164, 165]. It has been observed that algae could remove dye substances from Sewage efficiently. Mohan et al [151] studied that the alga Spirogyra was noticed to eliminate the azo dye reactive yellow 22 professionally. In the same way, Daneshvar et al [152] presented the elimination of malachite green via Cosmarium sp. In a further investigation by Sarwa et al[153] the microalga *Acutodesmus obliquus* was observed to eliminate acid red 66 azo dye from industrial waste water. In the same way, *Chlorella vulgaris* was discovered to eliminate Congo red with dye removal rate reached up to 83% at an initial concentration of 5 mg/L as well as 53% at a concentration of 25 mg/L [154]. Additionally , *Tetraselmis sp. and Nannochloropsi sp*., were obtained to be utilized in dye elimination from industrial waste waters [155].

5. Factors affecting adsorption industrial textile dyes onto zeolite and algae surfaces

5.1. Effect of contact time:

The dye removal% is high at the initial time of the adsorption process but it reduce still it reaches equilibrium, Armağan and Turan [166] had performed series of analyses against time (15, 30, 60, 120, 240, 360, 480, and 1200 min) at a steady color centralization of 25 mg/L and a strong/fluid proportion of 5%. The outcomes are given in Fig. 6 (a). As demonstrated in Fig. 6 (a), as the molding time builds, the measure of adsorption of receptive color into the zeolite increments fundamentally apparent that the majority of the adsorption happens inside the initial 2 h; yet when changes in pH and focus were considered, the blending time of 4 h was picked. The information got show that Everzol Yellow 3RS H/C, Everzol Black B, and Everzol Red 3BS were adsorbed into zeolite with recuperations (Ci-Cr/Ci) of 35%, 35%, and 25%, individually.In Figure 6 b shows the increment of percent expulsion of Malachite Green color by Ulva Lactuca as an adsorbent with expanding time, until harmony at around 90 min.[167-169]. Figure 6 c shows The impact of contact time on the adsorption of methylene blue for 0.01 g of adsorbent (Chitosan@ Zeolite nanocomposite) and introductory centralizations of 5 and 10 ppm at various occasions The outcomes show that with an increment in the process contact time, the adsorption will increment because of the increment in touch with the color particles with the adsorbent surface.[170] An increase in the adsorption happens by increasing the contact time to arrive at the balance time of around 240 minutes. As is seen, adsorption of color atoms at a high rate at first happens and afterward progressively diminishes until the adsorption onto the adsorbent ranges equilibrium.[171].According to Radoor, S., et al[158], through the initial time that the adsorption limit increments strongly however after 150 min it almost arrives at equilibrium [172] This is presumably in light of the fact that an enormous number of active sites are accessible in the initial phase of the adsorption technique that speeds up the limiting of CR to the layer. Notwithstanding, after some time as increasingly more color atoms are ingested onto the film surface, Though, over the long time as increasingly more color particles are retained onto the outside of the layer, the adsorption process turns out to be less favored. [173] It had been accounted for that increasing the contact time from 20 to 180 min, CR take-up capacity of PDA@ DCA-COOH film improves from 30 mg/g to 79.33 mg/g. Following 3 hours, no further improvement in adsorption limit was noticed, so the 3 hours were picked as the ideal time to direct adsorption studies.[174] While, The dye removal by *U. lactuca* as observed at different contact times and it was found that the adsorption capacity enhances with increasing in contact time up to about 2 h, after that time it is less stable [175]. The majority of the adsorption over adsorbent surfaces was completed after about 2 h. The dyes removal percentage from MB solution increase with increasing in contact time and is proved in available studies [176-179] As contact time increases, the percentage of elimination also rises in the beginning, but progressively comes close to a stable rate. These variations in the removal rate may be as the initial adsorption sites are accessible in addition to the initial dye concentration is elevated [176]. Also, table 3 shows the variation of contact time for various dyes adsorption on zeolites and algae .

5.2. Effect of concentration

Discharge of methylene blue color by means of natural and modified clinoptilolites utilizing color concentrations of 25–200 mg/l at the impartial pH has been appeared in Figure 7a. As needs be, the adsorption capacity of the colorant increased by increasing the initial concentration of the aquatic solution of the dye. In view of the outcomes, by increment of dye concentration, the efficiency of eliminating dye by the natural zeolite diminished from 67% to under 10%. Notwithstanding, increasing the initial concentration of the colorant didn't have any huge impacts on the process of dye elimination utilizing the magnetized adsorbent of the clinoptilolite. The discoveries showed that the colorant adsorption were 99.4%, 98.9%, 98.9%, 98.35%, 96%, and 95% at concentrations of 25, 50, 75, 100, 150, and 200 mg/l, individually. At the contact time of 5–90 min, the rate of decolorization by the natural zeolite elevated from 33% to 54.8% (Figure 7a). Additionally, 45% of the colorant was eliminated with at the contact time of Forty-Five minutes. By the by, in the wake of altering the adsorbent with iron oxide attractive nanoparticles at a similar contact time, the adsorption effectiveness came to 98.3%. Hence, the color expulsion measure moved toward the harmony condition after 45 min, which is the hour of balance of adsorption responses by the altered adsorbent.[180].

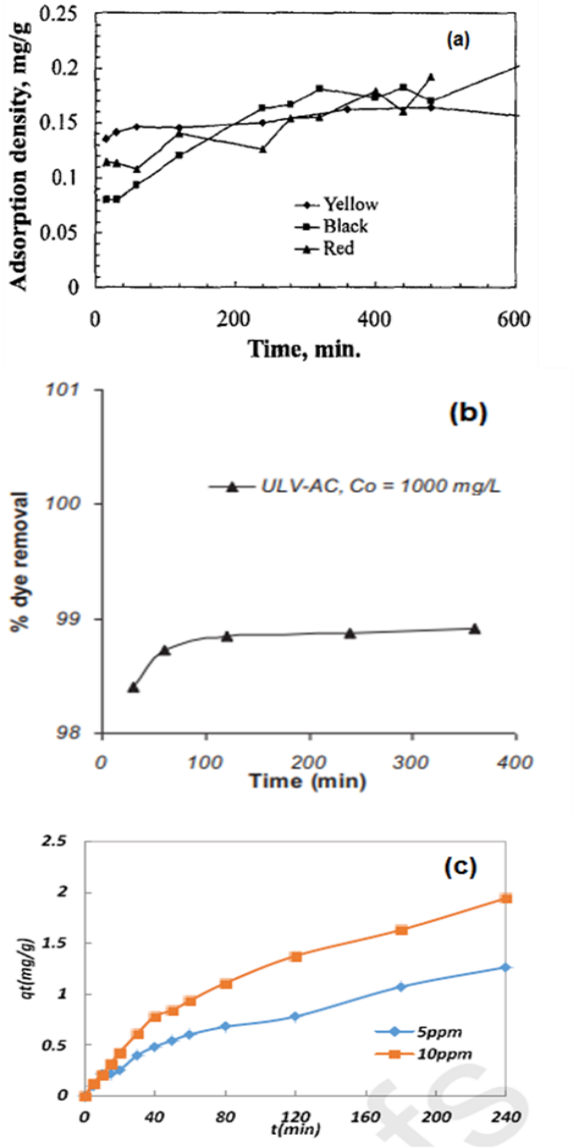

Figure 6. Effect of Contact time on the adsorption of :

(a) Everzol Yellow 3RS H/C, Everzol Black B and Everzol Red into zeolite with recoveries *(Ci-Cr/Ci)* of 35%, 35% and 25%, respectively. [166]

(b) Malachite Green dye into *Ulva lactuca* as an adsorbent with increasing time, until equilibrium at about 90 min [167]

(c) methylene blue for 0.01 g of adsorbent (Chitosan@ Zeolite nanocomposite) and initial concentrations of 5 and 10 ppm, equilibrium time of about 240 minutes [171]

Conversely, it has been found that the adsorption capacity of the adsorbent rises by increasing initial dye concentration of the solution and that's because by increasing the initial concentration due to overpowering the resistance of mass transfer, the momentum force is increased to transfer the mass between the adsorbent and the adsorbate [181] For instance, by increasing the initial concentration of Congo red dye in a range of (25-250) mg/L, the adsorption capacity of natural zeolite increased from 4.25 to 16.17 mg/g. Furthermore, as a result of the saturation of active sites, no considerable rise was noticed by rising initial concentration to 500 mg/L, and only adsorption capacity was 17.77 mg/g [182]. The effect of dye initial concentration on adsorption of Methylene Blue (MB) was investigated [175] over a spread of pH from 15 to 35 mg/L under constant parameter, pH 7, adsorbent dose 1.25 gr/L, contact time 110 min, and temperature 25 °C ± 2 °C. The solution was agitated continuously by using an orbital shaker. The effect of change of dye initial concentration at dye removal percentages is represented in Figure (7b). The result is in accordance with research conducted that supported in publish studied [183]. In Figure (7c), the impacts of initial concentration of Methyl Violet (MV) (10, 30 and 60 mg/L) and contact time (5–200 min) on the capacity of *P. sanctae-crucis* (PSC) brown algae bio-adsorbent are appeared at 298.15 K. According to the results mentioned, adsorption capacity enhanced from 4.776 mg/g to 26.463 mg/g since initial concentration of Methyl Violet (MV) dye raised from 10 mg/L to 60 mg/L, correspondingly. The tendency of rise in adsorption capacity was quick at initial times and the speed of adsorption method reduced as time passed; as active sites on the surface of the adsorbent are greater at initial times [184] and these sites are full by MV dye. It should also be mentioned that at The initial periods of contact time, over 80% of the initial concentration of 10 mg/L of the dye of the color being adsorbed by the adsorbent. As in Figure (7c), increase the initial concentration of the dye, the processed equilibrium time increased and this equilibrium time was determined at 20, 40 and 80 minutes of concentration of 10, 30 and 60 mg / L , respectively.[185]

Table 3: variation of contact time for diverse adsorption systems

Adsorbent	Adsorbate	Conditions	Maximum adsorption capacity (mg/g)	References
BA-CA	CV dye	Contact time: 2-240 min pH: 2-12 Adsorbent dosage: 0.1–2.0 g Temperature: 20°C Initial concentration: 20-400 mg/L	273.85 mg/g	[198]
BA	CV dye	Contact time: 2-240 min pH: 2-12 Adsorbent dosage: 0.1–2.0 g Temperature: 20°C Initial concentration: 20-400 mg/L	146.94 mg/g	[198]
Zeolite	Basic dye	Contact time: 0-180 min pH: 7 Adsorbent dosage: 2 g Temperature: 20°C Initial concentration: 50-500 mg/L	55.86	[199]
The brown alga	Methylene blue	Contact time: 0-400 min pH: 3-12 Adsorbent dosage: 0.01-0.2 g Temperature: 25, 35 and 45°C Initial concentration: 5, 20, 50 and 100 mg/L	38.61	[200]
Green alga	Methylene blue	Contact time: 5-120 min pH: 2-10 Adsorbent dosage: 0.1-1 g Temperature: 25 ± 2°C Initial concentration: 5-25 mg/L	40.2	[197]
Algae Gelidium	Methylene blue	Contact time: 0-190 min pH: 6 Temperature: 20°C Initial concentration: 40-800 mg/L	171	[201]
Green alga	Reactive red 5	Contact time: 0-360 min pH: 1-3 Temperature: 20-60°C Initial concentration: 200-240 mg/L	555.6	[202]

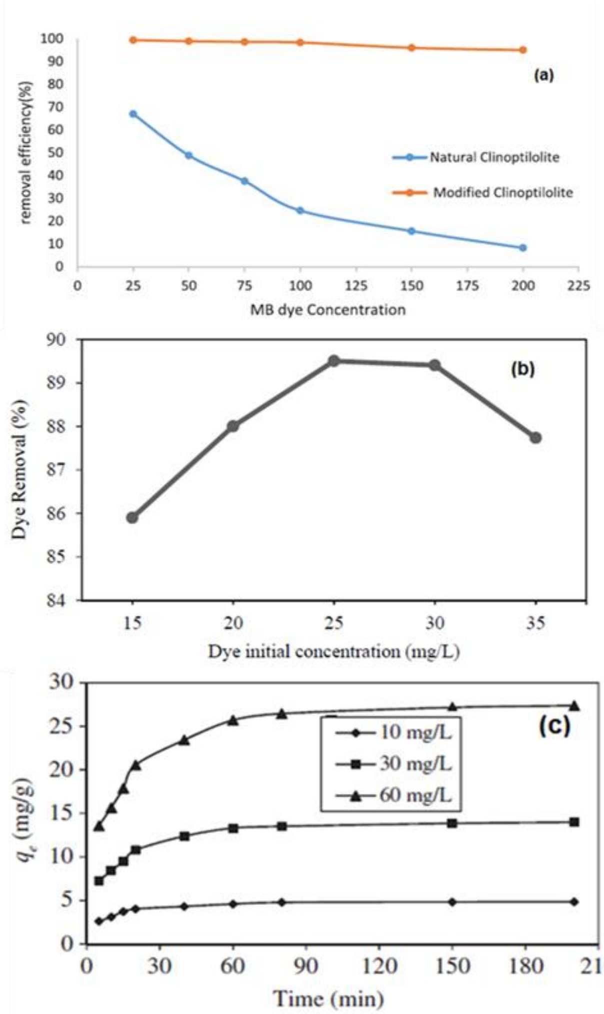

Figure 7. Effect of Concentration on:

(a) Removal of methylene blue dye by natural and modified clinoptilolites using dye concentrations of 25–200 mg/l at the neutral pH, contact time 45 min and adsorbent dose 0.5 gm.[180]
(b) Removal percentage of methylene blue dye by *Ulva lactuca*; over a spread of pH from 15 to 35 mg/L under constant parameter, pH 7, adsorbent dose 1.25 gr/L, contact time 110 min, and temperature 25 °C ± 2 °C.[175]
(c) Adsorption capacity of Methyl Violet MV dye using *P. sanctae-crucis* (PSC) brown algae (under the conditions of pH = 8, 2 g/L adsorbent dosage, temperature 25 °C, 150 rpm mixing rate).[185]

5.3. Effect of dose:

The effect of increasing the applied zeolite doses on the elimination percentages of safarnin molecules was investigated [186] and the achieved outcomes were in detail in Figure 8a For heulandite and clinoptilolite zeolite raw materials, the percentages presented obvious regular increase in the value through increasing the practical quantity from 0.05g to 0.15g realizing the greatest outcomes at 0.15g The dye elimination percentages were increased from 91.5% to 98.8% and from 88.5% to 95.4% for heulandite and clinoptilolite, correspondingly. This performance was documented through numerous authors and was explained to be commitment to the increase in the total of dynamic adsorption or receptor sites in addition to the whole surface areas of the reacting elements in the system [187, 188] On the other hand, elimination of safranin dye by clinoptilolite and heulandite was decreased notably once raising the used amount more than 0.02g, the elimination via phillipsite exhibited normal tendency with the increase of the used zeolite doses up to 0.25g (Fig. 8a). The detected values for phillipsite were increased by 44.2%, 53.2%, 68%, 85% and 88% with increased the useddoses by 0.05, 0.1, 0.15, 0.2 and 0.25g, correspondingly. The recorded reduction in the elimination values for clinoptiolite and heulandite with high doses over 0.02g was attributed to the reported decreasing in the pH of water in the existences of enormous quantities of heulandite group raw materials [189]. This can encourage the protonation methods and decrease the electrostatic attractions of such positive molecules [190, 191].

Additional investigation [192] has reported that The influence of adsorbent dose on the elimination of particular azo dyes was estimated by changing the amount of the modified zeolite, which varied from 0.05 g to 1 g, by maintaining stable initial dye concentration at 50.0 mg/L. As seen in Figure 8 b, elimination effectiveness of the reactive dyes enhanced through rising the quantity of adsorbent. It was noticed that above 93% adsorption was reached by utilizing 0.25 g of the sorbent and there was no considerable variation in dye elimination effectiveness. This could be as a result of the accessibility of additional adsorbent surface for the azo colorants to be adsorbed.

The influence of adsorbent amount on acid dyes elimination was estimated at 9 various values of this effective factor, in the variety from 1 g/L to 9 g/L, while the initial pH, dye concentration, temperature and contact time were saved stable at 2, 30 mg/L, 27 °C. and two hours, respectively, for all three dyes. As shown in Figure 8c, the adsorption effectiveness of completely 3 dyes gradually increased by increasing the adsorbent amount and reached highest values of 93.7% for Acid Blue 25 (AB25), 95.6% for Acid Orange 7 (AO7) and 87.1% for Acid Black 1 (AB1), at 2, 5 and 3 g, separately.[91] Such a performance, which is associated to the corresponding increase in the total of sites accessible for colorant adsorption, is in agreement with preceding explanations on Malachite Green elimination by various amounts for example *Pithophora sp.* [193], *Cosmarium sp*. and *Cladophora sp*. [194, 195] and Acid Black 1 biosorption by using *Cystoseira indica* and *Gracilaria persica* biomasses [196] Additional cause may be their dissimilar morphological and chemical structures. Conversely, the reduction in dyes elimination effectiveness noticed at adsorbent amount greater than the optimal values was suggested to be the outcome of increasing particle interaction and aggregation, causing a decrease of whole bio sorbent surface area[197, 198].

Furthermore, the effectiveness of malachite green (MG) colorant elimination increased by increasing the amount of adsorbent in case of utilizing marine algae *Enteromorpha*, in range 50-350 mg.[199]. With increasing the amount of *Caulerpa racemosa var. cylindracea*, a clear increase in the adsorbed colorant has been described.[200] Similar outcomes have been found for elimination through utilizing *Cystoseira barbatula[201]* as well as using *Chaetophora sp.[202]*. At lower quantities of the substance, the rate of elimination reduced due to the quick overload of adsorption sites with colorant ions. It has been observed that the percentage of elimination improved quickly while the quantity of adsorbent increased [203]. Methylene Blue (MB) adsorption capability increased by rising adsorbent quantity of *U. lactuca [177, 178]*.

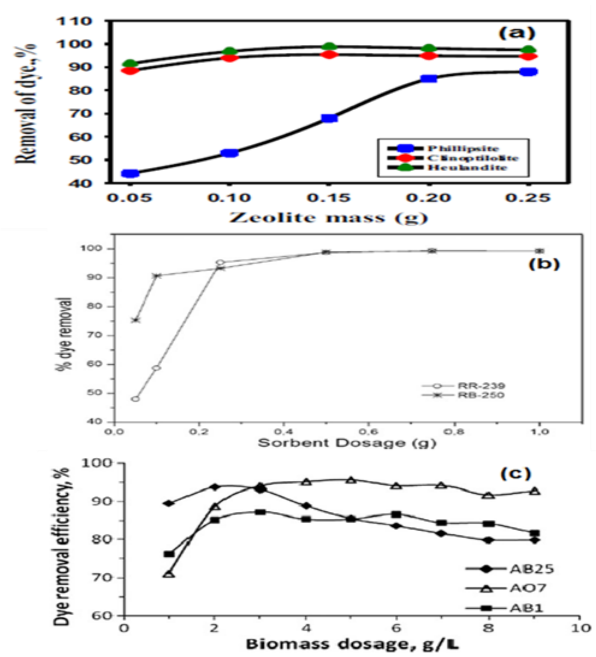

Figure 8. Effect of dose of :

(a) Heulandite, phillipsite and clinoptilolite zeolites masses on the removal percentages of safarnin dye.[186]
(b) Modified zeolite dosage on dye removal efficiency from single dye solutions; initial dye concentration: 50 mg/L; pH of dye solution: 7; temperature:20 °C.[192]
(c) Brown macro alga *Stoechospermum marginatum* on acidic dyes removal percentage. dose = 1.0 g ; concentration = 30 mg/L; pH = 2; T = 27 °C; where (AB25)= Acid Blue 25, (AO7) = Acid Orange 7 and (AB1)= Acid Black 1[91]

5.4. Effect of pH

The impact of pH on adsorption method plays a significant role as well as there is a certain pH value for each substance. The role of pH on the Congo red adsorption by PVA/SA/ZSM-5 zeolite membrane was investigated by changing the pH in range 3-11. It can be observed from Figure 4e that by increase in pH, the adsorption capacity of the membrane reduces from 5.33 to 2.92 mg/g. This could be because of variance in the surface charge of the membrane with pH. At small pH, the surface of membrane becomes protonated and consequently attracts anionic Congo red81. On the other hand, in basic pH the membrane surface tends to get negative and repel the anionic Congo red. Additionally, elevated pH strengthens the competition among Congo red colorant as well as hydroxide ions for the similar adsorption site.

The parameter of pH solution plays a significant role in colorant absorption investigations. A variation in the pH of the solution will modify the properties of both the colorant and the adsorbent. Consequently, adjusting the pH solution is an important factor for the adsorption system. The part of pH on the Congo red adsorption by using polyvinyl alcohol/sodium alginate/ZSM‑5 zeolite **(**PVA/SA/ZSM-5) zeolite membrane was investigated through changing the pH in range 3-11. As soon as pH solution increases, the absorption uptake of the composite reduces from 5.33 to 2.92 mg/g as presented in Figure 9 a [172]. This may be because of the variation in the surface charge of the composite beside the pH. At small pH, the protons of composite surface are dissolved and thus draw Congo red [204]. However, at higher pH, the surface of the membrane tends to become negative and ejects the anionic Congo Red. Additionally, higher pH improves the competition between CR and (OH^-) for the similar adsorption site.[172]. As These outcomes are according to research showed that supported in publish investigated [177, 179, 205] where an rising Methylene blue (MB) elimination rate gradually increased with pH solution. The colorant adsorption onto surface of biomass adsorbent is affected by ionic attractiveness [205] .This algal bio sorption has effectively been credited to the cell wall properties in spite of electrostatic attraction and complexion will play a an important role. Cell wall of algae is frequently including carbohydrates as well as protein that donate a functional group for example amino, hydroxyl, carboxyl, sulfate that plays as binding sites for metals [177, 178, 206] At smaller pH, fewer anionic adsorption site onto the surface of *U. lactuca* was formed, Additionally, sorption was negative, maybe because of extra protons competing with colorant molecule for active site on dried *U. lactuca* surface. The surface algal cell is responsible for binding colorant molecules [177].

In that investigation [205], the elimination rate of quick orange colorant at various selected pH are exhibited in Figure 9b. The outcomes present that the adsorption of colorant onto the surface of biomass is regulated by ionic attractiveness. When pH rate increased from 1 to 5, the adsorption uptake was improved expressively from 28.7 to 65.7% and then the colorant elimination rates were not appreciably changed beyond pH 5. Because pH reduced, the total of negative charge adsorbent sites reduced also the total of positive charge surface sites enhanced, which did not prefer the adsorption of positively charged colorant cations as a result of electrostatic revulsion. Similarly, less adsorption of fast orange at lower pH is because of the existence of extra protons competing with colorant positive ions for the adsorption sites. Parallel results were reported by several investigators [207-211] .

Additional report [212], The initial pH of colorants media not only influences the external charge of bio adsorbent, but also the rate of ionization of colorants may be changed. For studying the difference in the adsorption method with varying the pH of colorants solutions, Hence, the pH-dependence of the adsorption performance was investigated with varying the pH values of Crystal violet (CV) and Methylene blue (MB) contaminations from 3 to 9. As can be determined from the records Figure 9 c, the attractive *Fucus Vesiculosus* (Brown Algae) m-FV showed deficient sensitivity to the pH of both MB and CV solutions. It can be observed that in the wide-ranging of pHs, the ammonium pendants on colorants such as CV and MB exist in the cationic shape. Furthermore, the high separation performance of $–OSO_3$ - groups on FV causes the m-FV as a pH self-sufficient bio adsorbent, which present in the anionic shape at pH more than 3 [212]. Foroutan, Mohammadi et al.[213] investigated The influence of the initial pH from 2 to 9 on the colorants adsorption effectiveness on the surface of activated carbon of *Sargassum oligocystum*/Fe_3O_4 magnetic composite (ACSO/Fe_3O_4) then in accordance with Figure 9d, the elimination effectiveness of methyl violet MV in addition methylene blue MB enhanced with elevating pH, demonstrating that the extreme adsorption effectiveness was reached in neutral and basic pH values.[213]. In lower pH values, paralleled to basic one, the adsorption effectiveness of cationic colorants is smaller since in the acidic media the sum of protons is high as well as competes with positively charged colorant to select active sites onto the adsorbent also stops additional colorants adsorption [214]. Furthermore, in acidic environments, functional groups for instance carboxyl as well as Fe-OH have a positive charge (Fig. 10). Therefore, a repulsive electrostatic strength is created among the ACSO/Fe_3O_4 magnetic combinations and the colorant molecule and so the adsorption effectiveness is decreased [215, 216]. In spite of this repulsive strength in the acidic environments, at pH 2, MB along with MV molecules were adsorbed 62.43% and 67.53%, separately, by using ACSO/Fe_3O_4. This comparatively appropriate adsorption may be validated by the powerful π-π interactions among the surface of the ACSO/Fe_3O_4 magnetic combination and cationic colorants. Such outcomes have been described in previous investigations [217]

5.5. Effect of temperature

Temperature parameter negatively affects the colorant adsorption technique [218] but in another investigations didn't influence [182, 183] . So as to consider the influence of temperature besides the distinction nature and the practicability of Congo red (CR) adsorption into natural clinoptilolite (NC) and surfactant modified clinoptilolite SMC, the adsorption process was studied in temperature regulated shaking (Figure 11 a) presented that temperature didn't influence adsorption of CR on NC and SMC surfaces.[182]

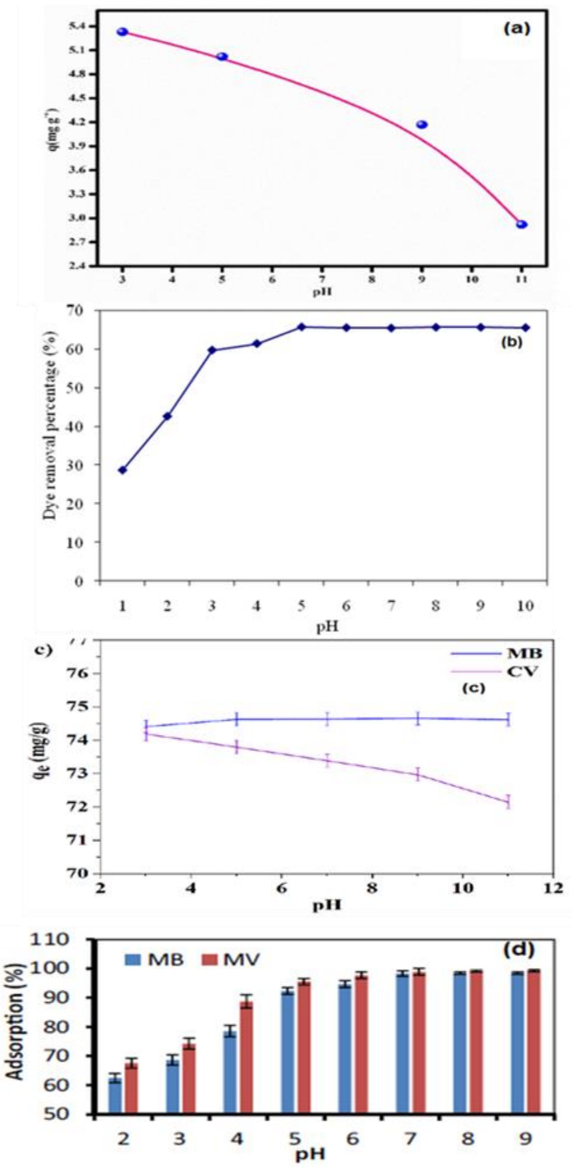

Figure 9. Effect of pH on :

(a) Adsorption of Congo red onto PVA/SA/ZSM-5 zeolite membrane (adsorbent dose = 2.5 wt%, initial CR concentration = 10 ppm, contact time = 130 min, pH = 3 and temperature = 30 °C).[172]

(b) Adsorption of dye by non-living biomass *Laurencia papillosa* (dye concentration=20 mg/l, contact time = 60 min adsorbent dose = 2 g, temperature= 25±2°C).[205]

(c) Adsorption capacity of m-FV for MB and CV (25 mL of 300 mg/g of MB; adsorbent dose: 25 mg; room temperature).

(d) The removal of methylene blue (MB) and methyl violet (MV) onto ACSO/Fe_3O_4 from aqueous solutions. (contact time: 50 min, Ci: 20 mg/L, ACSO/Fe_3O_4 dose: 0.15 g/100 mL, temperature of 25 °C.[213]

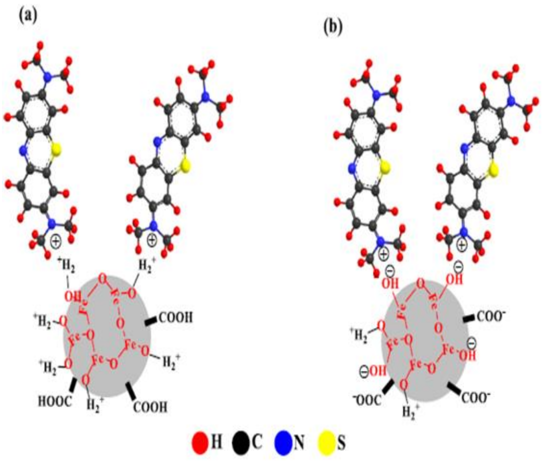

Figure 10, [213] The interactions between the ACSO/Fe_3O_4 and cationic dye at (a) Acidic pH (b) Alkaline pH

A further investigation [185] Temperature factor is considered as one of the significant factors in the adsorption method. It has a particular and specific influence on adsorption method as the capability of equilibrium adsorption of Methyl violet (MV) on the adsorbent changes by changing temperature [219]. Figure 11b exhibits the influence of temperature variations on adsorption effectiveness of MV. Adsorption effectiveness of MV at equilibrium time which is eighty minutes decreased from 95.52% to 90.22% since temperature increased from 298.15 K to 318.15 K. This exhibits that adsorption method of MV colorant from the aqueous media by utilizing brown algae *Padina sanctae-crucis* (PSC) is exothermic which is regular in physical adsorption methods. As a result, reduction in the effectiveness of the adsorption because of elevated temperatures may be reserved to variations in active sites of the adsorbent as well as the tendency of the adsorbed substance to become away from the active sites in the aqueous solution. In earlier investigations, parallel outcomes were reported that effectiveness as well as adsorption capability of the colorant reduced as temperature reduced [218]

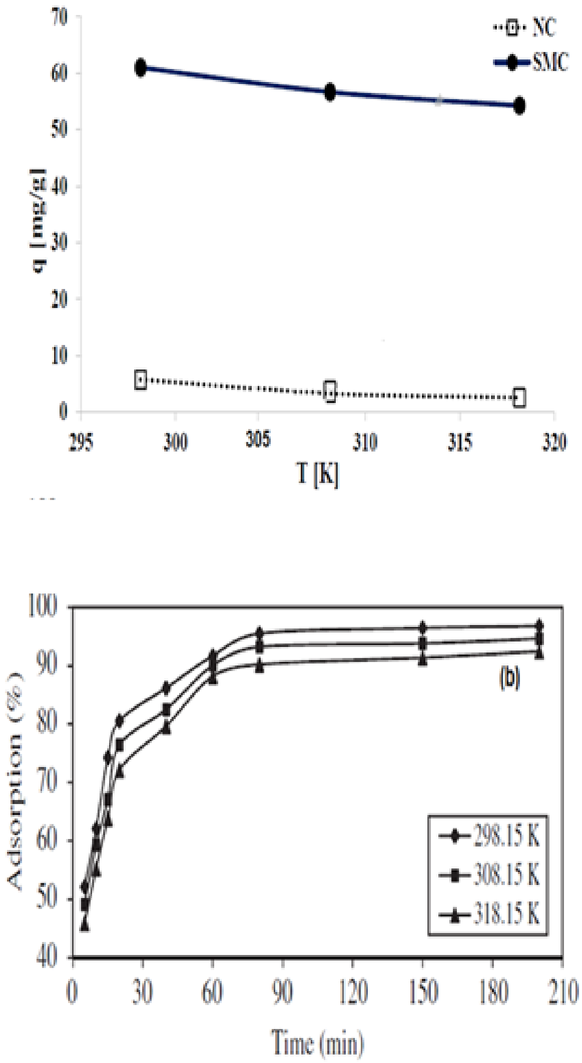

Figure 11. Effect of Temperature on :

(a) CR dye removal for natural clinoptillolite NC and surfactant modified clinoptilolite SMC.[182]

(b) MV dye removal using PSC (pH = 8, 2 g/L adsorbent dosage, 10 mg/L initial [185]

Conclusion:

In this review, we present definition and classification of marine algae, definition of Zeolite, industrial wastewater and their effect on the environment, water treatment technologies which involve chemical, biological, Combinatorial method, nanotechnology-based and physical methods, applications of algae and natural zeolite in wastewater treatment separately and in combination. Finally, we discuss the factors that affect dyes adsorption onto zeolite and marine algae surfaces, such as contact time, temperature, solution pH and catalyst dose. And concluded that the dye removal percentage is high at the first minutes of the adsorption process but it reduces gradually still it reaches equilibrium, Temperature negatively affects the dye adsorption method, There is a specific pH value for each catalyst, at which the optimum adsorption of dyes happens and adsorbent dose growth in general enhances catalytic activity because of the increase in total surface area and the total of active sites on catalyst surface. The factors that affect dye adsorption onto zeolite and marine algae surfaces should be known, for getting optimum conditions for dye adsorption onto zeolite and marine algae.

References:-

[1] Alvarez PJ, Chan CK, Elimelech M, Halas NJ, Villagrán D. Emerging opportunities for nanotechnology to enhance water security. Nature nanotechnology 2018;13:634.

[2] Mauter MS, Zucker I, Perreault F, Werber JR, Kim J-H, Elimelech M. The role of nanotechnology in tackling global water challenges. Nature Sustainability 2018;1:166-75.

[3] Yang X, Wang Z, Shao L. Construction of oil-unidirectional membrane for integrated oil collection with lossless transportation and oil-in-water emulsion purification. Journal of Membrane Science 2018;549:67-74.

[4] Pronk W, Ding A, Morgenroth E, Derlon N, Desmond P, Burkhardt M, et al. Gravity-driven membrane filtration for water and wastewater treatment: a review. Water research 2019;149:553-65.

[5] Fan H, Gu J, Meng H, Knebel A, Caro J. High-flux membranes based on the covalent organic framework COF-LZU1 for selective dye separation by nanofiltration. Angewandte Chemie International Edition 2018;57:4083-7.

[6] Zhang Y, Yu W, Li R, Xu Y, Shen L, Lin H, et al. Novel conductive membranes breaking through the selectivity-permeability trade-off for Congo red removal. Separation and Purification Technology 2019;211:368-76.

[7] Xu Y, Tognia M, Guo D, Shen L, Li R, Lin H. Facile preparation of polyacrylonitrile-co-methylacrylate based integrally skinned asymmetric nanofiltration membranes for sustainable molecular separation: An one-step method. Journal of colloid and interface science 2019;546:251-61.

[8] Banerjee S, Sharma GC, Chattopadhyaya M, Sharma YC. Kinetic and equilibrium modeling for the adsorptive removal of methylene blue from aqueous solutions on of activated fly ash (AFSH). Journal of Environmental Chemical Engineering 2014;2:1870-80.

[9] Aravind P, Subramanyan V, Ferro S, Gopalakrishnan R. Eco-friendly and facile integrated biological-cum-photo assisted electrooxidation process for degradation of textile wastewater. Water Research 2016;93:230-41.

[10] Zhou L, Zhou H, Yang X. Preparation and performance of a novel starch-based inorganic/organic composite coagulant for textile wastewater treatment. Separation and Purification Technology 2019;210:93-9.

[11] Khatri J, Nidheesh P, Singh TA, Kumar MS. Advanced oxidation processes based on zero-valent aluminium for treating textile wastewater. Chemical Engineering Journal 2018;348:67-73.

[12] Kumar PS, Varjani SJ, Suganya S. Treatment of dye wastewater using an ultrasonic aided nanoparticle stacked activated carbon: kinetic and isotherm modelling. Bioresource technology 2018;250:716-22.

[13] Unuabonah EI, Taubert A. Clay–polymer nanocomposites (CPNs): Adsorbents of the future for water treatment. Applied clay science 2014;99:83-92.

[14] Bi B, Xu L, Xu B, Liu X. Heteropoly blue-intercalated layered double hydroxides for cationic dye removal from aqueous media. Applied Clay Science 2011;54:242-7.

[15] Villegas LGC, Mashhadi N, Chen M, Mukherjee D, Taylor KE, Biswas N. A short review of techniques for phenol removal from wastewater. Current Pollution Reports 2016;2:157-67.

[16] Chapman AC, Siebold A. On the application of adsorption to the detection and separation of certain dyes. Analyst 1912;37:339-45.

[17] رحمانی م, ساسانی م. Evaluation of 3A zeolite as an adsorbent for the decolorization of rhodamine B dye in contaminated waters. شیمی کاربردی 90-11:83;2016.

[18] Habiba U, Siddique TA, Joo TC, Salleh A, Ang BC, Afifi AM. Synthesis of chitosan/polyvinyl alcohol/zeolite composite for removal of methyl orange, Congo red and chromium (VI) by flocculation/adsorption. Carbohydrate polymers 2017;157:1568-76.

[19] McKay G, Porter J, Prasad G. The removal of dye colours from aqueous solutions by adsorption on low-cost materials. Water, Air, and Soil Pollution 1999;114:423-38.

[20] El-Naggar NE-A, Hamouda RA, Saddiq AA, Alkinani MH. Simultaneous bioremediation of cationic copper ions and anionic methyl orange azo dye by brown marine alga Fucus vesiculosus. Scientific Reports 2021;11:1-19.

[21] Marungrueng K, Pavasant P. High performance biosorbent (Caulerpa lentillifera) for basic dye removal. Bioresource technology 2007;98:1567-72.

[22] Kim S-K, Thomas NV, Li X. Anticancer compounds from marine macroalgae and their application as medicinal foods. Advances in

food and nutrition research: Elsevier; 2011. p. 213-24.
[23] Na H-J, Moon P-D, Lee H-J, Kim H-R, Chae H-J, Shin T, et al. Regulatory effect of atopic allergic reaction by Carpopeltis affinis. Journal of ethnopharmacology 2005;101:43-8.
[24] Bouhlal R, Haslin C, Chermann J-C, Colliec-Jouault S, Sinquin C, Simon G, et al. Antiviral activities of sulfated polysaccharides isolated from Sphaerococcus coronopifolius (Rhodophytha, Gigartinales) and Boergeseniella thuyoides (Rhodophyta, Ceramiales). Marine drugs 2011;9:1187-209.
[25] Kim S-K, Karadeniz F. Anti-HIV activity of extracts and compounds from marine algae. Advances in food and nutrition research: Elsevier; 2011. p. 255-65.
[26] Shi D, Li J, Guo S, Han L. Antithrombotic effect of bromophenol, the alga-derived thrombin inhibitor. 13th International Biotechnology Symposium and Exhibition (第 13 届 IUPAC 国际生物工程会议): 大连理工大学; 2008. p. 579-.
[27] de Felício R, de Albuquerque S, Young MCM, Yokoya NS, Debonsi HM. Trypanocidal, leishmanicidal and antifungal potential from marine red alga Bostrychia tenella J. Agardh (Rhodomelaceae, Ceramiales). Journal of Pharmaceutical and Biomedical Analysis 2010;52:763-9.
[28] Devi GK, Manivannan K, Thirumaran G, Rajathi FAA, Anantharaman P. In vitro antioxidant activities of selected seaweeds from Southeast coast of India. Asian Pacific journal of tropical medicine 2011;4:205-11.
[29] Bhadury P, Wright PC. Exploitation of marine algae: biogenic compounds for potential antifouling applications. Planta 2004;219:561-78.
[30] Centeno POR, Ballantine DL. Effects of culture conditions on production of antibiotically active metabolites by the marine alga Spyridia filamentosa (Ceramiaceae, Rhodophyta). I. Light. Journal of applied phycology 1998;10:453.
[31] Grover P, Singh SK, Suri K, Bansal G. Phytochemical investigations and systematic exploration of anti-cancer potential of leaves of Nyctanthes arbor-tristis. Medicinal Plants-International Journal of Phytomedicines and Related Industries 2015;7:227-32.
[32] Mehl-Madrona L. Narrative medicine: The use of history and story in the healing process: Simon and Schuster; 2007.
[33] Singh S, Kate BN, Banerjee U. Bioactive compounds from cyanobacteria and microalgae: an overview. Critical reviews in biotechnology 2005;25:73-95.
[34] Pulz O, Gross W. Valuable products from biotechnology of microalgae. Applied microbiology and biotechnology 2004;65:635-48.
[35] Proksch P, Edrada R, Ebel R. Drugs from the seas–current status and microbiological implications. Applied microbiology and biotechnology 2002;59:125-34.
[36] El Gamal AA. Biological importance of marine algae. Saudi pharmaceutical journal 2010;18:1-25.
[37] Anbuchezhian R, Karuppiah V, Li Z. Prospect of marine algae for production of industrially important chemicals. Algal Biorefinery: An Integrated Approach: Springer; 2015. p. 195-217.
[38] Hillison J. The Concerns of Agricultural Education Pre-Service Students and First Year Teachers. Journal of the American Association of Teacher Educators in Agriculture 1977;18:33-40.
[39] Garson M. Biosynthetic studies on marine natural products. Natural Product Reports 1989;6:143-70.
[40] Margeta K, Logar NZ, Šiljeg M, Farkaš A. Natural zeolites in water treatment–how effective is their use. Water treatment 2013;5:81-112.
[41] Wajima T, Haga M, Kuzawa K, Ishimoto H, Tamada O, Ito K, et al. Zeolite synthesis from paper sludge ash at low temperature (90 C) with addition of diatomite. Journal of hazardous materials 2006;132:244-52.
[42] Filippidis A, Kantiranis N. Experimental neutralization of lake and stream waters from N. Greece using domestic HEU-type rich natural zeolitic material. Desalination 2007;213:47-55.
[43] Al-Moameri H, Zhao Y, Ghoreishi R, Suppes GJ. Simulation blowing agent performance, cell morphology, and cell pressure in rigid polyurethane foams. Industrial & Engineering Chemistry Research 2016;55:2336-44.
[44] Mumpton FA. La roca magica: Uses of natural zeolites in agriculture and industry. Proceedings of the National Academy of Sciences 1999;96:3463-70.
[45] Seow TW, Lim CK, Nor MHM, Mubarak M, Lam CY, Yahya A, et al. Review on wastewater treatment technologies. Int J Appl Environ Sci 2016;11:111-26.
[46] Sharma B, Dangi AK, Shukla P. Contemporary enzyme based technologies for bioremediation: a review. Journal of environmental management 2018;210:10-22.
[47] Khan S, Malik A. Toxicity evaluation of textile effluents and role of native soil bacterium in biodegradation of a textile dye. Environmental Science and Pollution Research 2018;25:4446-58.

[48] Vikrant K, Giri BS, Raza N, Roy K, Kim K-H, Rai BN, et al. Recent advancements in bioremediation of dye: current status and challenges. Bioresource technology 2018;253:355-67.
[49] Ito T, Adachi Y, Yamanashi Y, Shimada Y. Long–term natural remediation process in textile dye–polluted river sediment driven by bacterial community changes. Water research 2016;100:458-65.
[50] Gupta VK, Ali I, Saleh TA, Nayak A, Agarwal S. Chemical treatment technologies for waste-water recycling—an overview. Rsc Advances 2012;2:6380-8.
[51] Periyasamy AP, Ramamoorthy SK, Rwawiire S, Zhao Y. Sustainable wastewater treatment methods for textile industry. Sustainable Innovations in Apparel Production: Springer; 2018. p. 21-87.
[52] Patel H, Vashi R. Characterization and treatment of textile wastewater: Elsevier; 2015.
[53] Ghaly A, Ananthashankar R, Alhattab M, Ramakrishnan V. Production, characterization and treatment of textile effluents: a critical review. J Chem Eng Process Technol 2014;5:1-18.
[54] Quach-Cu J, Herrera-Lynch B, Marciniak C, Adams S, Simmerman A, Reinke RA. The effect of primary, secondary, and tertiary wastewater treatment processes on antibiotic resistance gene (ARG) concentrations in solid and dissolved wastewater fractions. Water 2018;10:37.
[55] Yin H, Qiu P, Qian Y, Kong Z, Zheng X, Tang Z, et al. Textile wastewater treatment for water reuse: a case study. Processes 2019;7:34.
[56] Bharagava RN, Chowdhary P. Emerging and eco-friendly approaches for waste management: Springer; 2019.
[57] Al Prol AE. Study of environmental concerns of dyes and recent textile effluents treatment technology: A Review. Asian Journal of Fisheries and Aquatic Research 2019:1-18.
[58] Crini G, Lichtfouse E. Advantages and disadvantages of techniques used for wastewater treatment. Environmental Chemistry Letters 2019;17:145-55.
[59] Saratale RG, Saratale GD, Chang J-S, Govindwar S. Bacterial decolorization and degradation of azo dyes: a review. Journal of the Taiwan Institute of Chemical Engineers 2011;42:138-57.
[60] Morató J, Carneiro AP, Ortiz A, Gallegos Á. Sustainable technologies for water treatment. What is Sustainable Technology?: Routledge; 2017. p. 190-212.
[61] Neoh CH, Noor ZZ, Mutamim NSA, Lim CK. Green technology in wastewater treatment technologies: integration of membrane bioreactor with various wastewater treatment systems. Chemical Engineering Journal 2016;283:582-94.
[62] Roy M, Saha R. Dyes and their removal technologies from wastewater: A critical review. Intelligent Environmental Data Monitoring for Pollution Management 2021:127-60.
[63] Mani S, Chowdhary P, Bharagava RN. Textile wastewater dyes: toxicity profile and treatment approaches. Emerging and eco-friendly approaches for waste management: Springer; 2019. p. 219-44.
[64] Holkar CR, Jadhav AJ, Pinjari DV, Mahamuni NM, Pandit AB. A critical review on textile wastewater treatments: possible approaches. Journal of environmental management 2016;182:351-66.
[65] Al-Amin M, Dey SC, Rashid TU, Ashaduzzaman M, Shamsuddin SM. Solar assisted photocatalytic degradation of reactive azo dyes in presence of anatase titanium dioxide. International Journal of Latest Research in Engineering and Technology 2016;2:14-21.
[66] Lam S-M, Low X-ZD, Wong K-A, Sin J-C. Sequencing coagulation–photodegradation treatment of Malachite Green dye and textile wastewater through ZnO micro/nanoflowers. Chemical Engineering Communications 2018;205:1143-56.
[67] Ghoreishian SM, Badii K, Norouzi M, Rashidi A, Montazer M, Sadeghi M, et al. Decolorization and mineralization of an azo reactive dye using loaded nano-photocatalysts on spacer fabric: kinetic study and operational factors. Journal of the Taiwan Institute of Chemical Engineers 2014;45:2436-46.
[68] Bougdour N, Tiskatine R, Bakas I, Assabbane A. PHotocatalytic degradation of industrial textile wastewater using S2O82−/Fe2+ process. Materials Today: Proceedings 2020;22:69-72.
[69] Bouras HD, Isik Z, Arikan EB, Bouras N, Chergui A, Yatmaz HC, et al. Photocatalytic oxidation of azo dye solutions by impregnation of ZnO on fungi. Biochemical engineering journal 2019;146:150-9.
[70] Ayare SD, Gogate PR. Sonocatalytic treatment of phosphonate containing industrial wastewater intensified using combined oxidation approaches. Ultrasonics sonochemistry 2019;51:69-76.
[71] Jorfi S, Pourfadakari S, Kakavandi B. A new approach in sono-photocatalytic degradation of recalcitrant textile wastewater using MgO@ Zeolite nanostructure under UVA irradiation. Chemical Engineering Journal 2018;343:95-107.
[72] Jothirani R, Kumar PS, Saravanan A, Narayan AS, Dutta A. Ultrasonic modified corn pith for the sequestration of dye from aqueous solution.

Journal of Industrial and Engineering Chemistry 2016;39:162-75.
[73] Hannan MA, Haque P, Kabir SF, Rahman MM. Scope of sustainable pretreatment of cotton knit fabric avoiding major chemicals. Journal of Natural Fibers 2018.
[74] Purkait M, DasGupta S, De S. Removal of dye from wastewater using micellar-enhanced ultrafiltration and recovery of surfactant. Separation and purification Technology 2004;37:81-92.
[75] Anirudhan T, Ramachandran M. Adsorptive removal of basic dyes from aqueous solutions by surfactant modified bentonite clay (organoclay): kinetic and competitive adsorption isotherm. Process Safety and Environmental Protection 2015;95:215-25.
[76] Yi S, Sun G, Dai F. Removal and separation of mixed ionic dyes by solvent extraction. Textile research journal 2018;88:1641-9.
[77] Yi S, Sun S, Dai F. Removal, separation, and recovery of mixed ionic dyes by solvent extraction using reverse micellar systems. Chemistry Letters 2015;44:1173-5.
[78] Levin L, Papinutti L, Forchiassin F. Evaluation of Argentinean white rot fungi for their ability to produce lignin-modifying enzymes and decolorize industrial dyes. Bioresource Technology 2004;94:169-76.
[79] Panizza M, Cerisola G. Electrocatalytic materials for the electrochemical oxidation of synthetic dyes. Applied Catalysis B: Environmental 2007;75:95-101.
[80] Wang A, Li YY, Ru J. The mechanism and application of the electro-Fenton process for azo dye Acid Red 14 degradation using an activated carbon fibre felt cathode. Journal of Chemical Technology & Biotechnology 2010;85:1463-70.
[81] Forgacs E, Cserhati T, Oros G. Removal of synthetic dyes from wastewaters: a review. Environment international 2004;30:953-71.
[82] Jasińska A, Różalska S, Bernat P, Paraszkiewicz K, Długoński J. Malachite green decolorization by non-basidiomycete filamentous fungi of Penicillium pinophilum and Myrothecium roridum. International biodeterioration & biodegradation 2012;73:33-40.
[83] Jayasinghe C, Imtiaj A, Lee GW, Im KH, Hur H, Lee MW, et al. Degradation of three aromatic dyes by white rot fungi and the production of ligninolytic enzymes. Mycobiology 2008;36:114-20.
[84] Jin X-C, Liu G-Q, Xu Z-H, Tao W-Y. Decolorization of a dye industry effluent by Aspergillus fumigatus XC6. Applied microbiology and biotechnology 2007;74:239-43.
[85] Rajesh KR, Rajasimman M, Rajamohan N, Sivaprakash B. Equilibrium and kinetic studies on sorption of malachite green using Hydrilla verticillata biomass. 2010.
[86] Aleksić M, Kušić H, Koprivanac N, Leszczynska D, Božić AL. Heterogeneous Fenton type processes for the degradation of organic dye pollutant in water—The application of zeolite assisted AOPs. Desalination 2010;257:22-9.
[87] Hoigne J. Mechanisms, rates and selectivities of oxidations of organic compounds initiated by ozonation of water. Handbook of Ozone Technology and Applications 1982;1.
[88] Oliveira FH, Osugi ME, Paschoal FM, Profeti D, Olivi P, Zanoni MVB. Electrochemical oxidation of an acid dye by active chlorine generated using Ti/Sn (1− x) Ir x O 2 electrodes. Journal of Applied Electrochemistry 2007;37:583-92.
[89] Gottschalk C, Libra J, Saupe A. Ozone in overview. Ozonation of water and waste water: a practical guide to understanding ozone and its application Wiley-Vch, Weinheim, Germany 2000:5-35.
[90] Cho BP, Yang T, Blankenship LR, Moody JD, Churchwell M, Beland FA, et al. Synthesis and characterization of N-demethylated metabolites of malachite green and leucomalachite green. Chemical research in toxicology 2003;16:285-94.
[91] Daneshvar E, Kousha M, Sohrabi MS, Khataee A, Converti A. Biosorption of three acid dyes by the brown macroalga Stoechospermum marginatum: isotherm, kinetic and thermodynamic studies. Chemical Engineering Journal 2012;195:297-306.
[92] Crini G. Non-conventional low-cost adsorbents for dye removal: a review. Bioresource technology 2006;97:1061-85.
[93] Forgiarini E, de Souza AAU. Toxicity of textile dyes and their degradation by the enzyme horseradish peroxidase (HRP). Journal of Hazardous Materials 2007;147:1073-8.
[94] Bhatia D, Sharma NR, Singh J, Kanwar RS. Biological methods for textile dye removal from wastewater: A review. Critical Reviews in Environmental Science and Technology 2017;47:1836-76.
[95] Nagai M, Sato T, Watanabe H, Saito K, Kawata M, Enei H. Purification and characterization of an extracellular laccase from the edible mushroom Lentinula edodes, and decolorization of chemically different dyes. Applied microbiology and biotechnology 2002;60:327-35.
[96] Abd El-Rahim WM, Moawad H. Enhancing bioremoval of textile dyes by eight fungal strains from media supplemented with gelatine wastes and sucrose. Journal of Basic Microbiology: An International Journal on Biochemistry,

Physiology, Genetics, Morphology, and Ecology of Microorganisms 2003;43:367-75.
[97] Acuner E, Dilek F. Treatment of tectilon yellow 2G by Chlorella vulgaris. Process Biochemistry 2004;39:623-31.
[98] Aguilar M, Saez J, Llorens M, Soler A, Ortuno J. Nutrient removal and sludge production in the coagulation–flocculation process. Water research 2002;36:2910-9.
[99] Aguilar M, Sáez J, Lloréns M, Soler A, Ortuño JF, Meseguer V, et al. Improvement of coagulation–flocculation process using anionic polyacrylamide as coagulant aid. Chemosphere 2005;58:47-56.
[100] Aksu Z. Application of biosorption for the removal of organic pollutants: a review. Process biochemistry 2005;40:997-1026.
[101] Jinqi L, Houtian L. Degradation of azo dyes by algae. Environmental pollution 1992;75:273-8.
[102] Kamel M, El-Shishtawy RM, Yussef B, Mashaly H. Ultrasonic assisted dyeing: III. Dyeing of wool with lac as a natural dye. Dyes and pigments 2005;65:103-10.
[103] Joshi M, Bansal R, Purwar R. Colour removal from textile effluents. 2004.
[104] Ahmad A, Mohd-Setapar SH, Chuong CS, Khatoon A, Wani WA, Kumar R, et al. Recent advances in new generation dye removal technologies: novel search for approaches to reprocess wastewater. RSC advances 2015;5:30801-18.
[105] Wu Z-C, Zhang Y, Tao T-X, Zhang L, Fong H. Silver nanoparticles on amidoxime fibers for photo-catalytic degradation of organic dyes in waste water. Applied Surface Science 2010;257:1092-7.
[106] Arunachalam R, Dhanasingh S, Kalimuthu B, Uthirappan M, Rose C, Mandal AB. Phytosynthesis of silver nanoparticles using Coccinia grandis leaf extract and its application in the photocatalytic degradation. Colloids and Surfaces B: Biointerfaces 2012;94:226-30.
[107] Suvith V, Philip D. Catalytic degradation of methylene blue using biosynthesized gold and silver nanoparticles. Spectrochimica Acta Part A: Molecular and Biomolecular Spectroscopy 2014;118:526-32.
[108] Biswas S, Rashid TU, Debnath T, Haque P, Rahman MM. Application of chitosan-clay biocomposite beads for removal of heavy metal and dye from industrial effluent. Journal of Composites Science 2020;4:16.
[109] Bhuiyan MR, Rahman MM, Shaid A, Khan M. Decolorization of textile wastewater by gamma irradiation and its reuse in dyeing process. Desalination and Water Treatment 2015;54:2848-55.
[110] Jiang M, Ye K, Deng J, Lin J, Ye W, Zhao S, et al. Conventional ultrafiltration as effective strategy for dye/salt fractionation in textile wastewater treatment. Environmental science & technology 2018;52:10698-708.
[111] Ma X, Chen P, Zhou M, Zhong Z, Zhang F, Xing W. Tight ultrafiltration ceramic membrane for separation of dyes and mixed salts (both NaCl/Na2SO4) in textile wastewater treatment. Industrial & Engineering Chemistry Research 2017;56:7070-9.
[112] Royer B, Cardoso NF, Lima EC, Macedo TR, Airoldi C. A useful organofunctionalized layered silicate for textile dye removal. Journal of hazardous materials 2010;181:366-74.
[113] Dotto G, Esquerdo V, Vieira M, Pinto L. Optimization and kinetic analysis of food dyes biosorption by Spirulina platensis. Colloids and Surfaces B: Biointerfaces 2012;91:234-41.
[114] Eren Z, Ince NH. Sonolytic and sonocatalytic degradation of azo dyes by low and high frequency ultrasound. Journal of hazardous materials 2010;177:1019-24.
[115] Wang S, Ng CW, Wang W, Li Q, Li L. A comparative study on the adsorption of acid and reactive dyes on multiwall carbon nanotubes in single and binary dye systems. Journal of Chemical & Engineering Data 2012;57:1563-9.
[116] Bhattacharya P, Dutta S, Ghosh S, Vedajnananda S, Bandyopadhyay S. Crossflow microfiltration using ceramic membrane for treatment of sulphur black effluent from garment processing industry. Desalination 2010;261:67-72.
[117] Cheriaa J, Bakhrouf A. Triphenylmethanes, malachite green and crystal violet dyes decolourisation by Sphingomonas paucimobilis. Annals of microbiology 2009;59:57-61.
[118] Eichlerová I, Homolka L, Nerud F. Ability of industrial dyes decolorization and ligninolytic enzymes production by different Pleurotus species with special attention on Pleurotus calyptratus, strain CCBAS 461. Process Biochemistry 2006;41:941-6.
[119] Fu Y, Viraraghavan T. Fungal decolorization of dye wastewaters: a review. Bioresource technology 2001;79:251-62.
[120] Fu Y, Viraraghavan T. Removal of Congo Red from an aqueous solution by fungus Aspergillus niger. Advances in Environmental Research 2002;7:239-47.
[121] Fu Y, Viraraghavan T. Dye biosorption sites in Aspergillus niger. Bioresource technology 2002;82:139-45.
[122] Fu Y, Viraraghavan T. Column studies for biosorption of dyes from aqueous solutions on immobilised Aspergillus niger fungal biomass. Water Sa 2003;29:465-72.

[123] Galhaup C, Wagner H, Hinterstoisser B, Haltrich D. Increased production of laccase by the wood-degrading basidiomycete Trametes pubescens. Enzyme and Microbial Technology 2002;30:529-36.
[124] Eichlerova I, Homolka L, Nerud F. Evaluation of synthetic dye decolorization capacity in Ischnoderma resinosum. Journal of Industrial Microbiology and Biotechnology 2006;33:759-66.
[125] Eichlerová I, Homolka L, Nerud F. Synthetic dye decolorization capacity of white rot fungus Dichomitus squalens. Bioresource Technology 2006;97:2153-9.
[126] Eichlerova I, Homolka L, Nerud F. Decolorization of high concentrations of synthetic dyes by the white rot fungus Bjerkandera adusta strain CCBAS 232. Dyes and Pigments 2007;75:38-44.
[127] El-Aty A, Mostafa FA. Effect of various media and supplements on laccase activity and its application in dyes decolorization. Malaysian Journal of Microbiology 2013;9:166-75.
[128] Eshghi H, Alishahi Z, Zokaei M, Daroodi A, Tabasi E. Decolorization of methylene blue by new fungus: Trichaptum biforme and decolorization of three synthetic dyes by Trametes hirsuta and Trametes gibbosa. European Journal of Chemistry 2011;2:463-8.
[129] Faraco V, Pezzella C, Miele A, Giardina P, Sannia G. Bio-remediation of colored industrial wastewaters by the white-rot fungi Phanerochaete chrysosporium and Pleurotus ostreatus and their enzymes. Biodegradation 2009;20:209-20.
[130] Fu Y, Viraraghavan T. Removal of a dye from an aqueous solution by the fungus Aspergillus niger. Water Quality Research Journal 2000;35:95-112.
[131] Pedersen AJ. Characterization and electrodialytic treatment of wood combustion fly ash for the removal of cadmium. Biomass and Bioenergy 2003;25:447-58.
[132] Oztekin E, Altin S. Wastewater treatment by electrodialysis system and fouling problems. Turkish Online Journal of Science & Technology 2016;6.
[133] Chang D, Choo K, Jung J, Jiang L, Ahn J, Nam M, et al. Foulant identification and fouling control with iron oxide adsorption in electrodialysis for the desalination of secondary effluent. Desalination 2009;236:152-9.
[134] Chen G. Electrochemical technologies in wastewater treatment. Separation and purification Technology 2004;38:11-41.
[135] Mohammadi T, Razmi A, Sadrzadeh M. Effect of operating parameters on Pb2+ separation from wastewater using electrodialysis. Desalination 2004;167:379-85.
[136] Lee H-J, Moon S-H, Tsai S-P. Effects of pulsed electric fields on membrane fouling in electrodialysis of NaCl solution containing humate. Separation and Purification Technology 2002;27:89-95.
[137] Shah MP. Azo dye removal technologies. Advances in research and applications 2018:169.
[138] Gao H, Chu X, Wang Y, Zhou F, Zhao K, Mu Z, et al. Media optimization for laccase production by Trichoderma harzianum ZF-2 using response surface methodology. Journal of microbiology and biotechnology 2013;23:1757-64.
[139] Fenoradosoa TA, Ali G, Delattre C, Laroche C, Petit E, Wadouachi A, et al. Extraction and characterization of an alginate from the brown seaweed Sargassum turbinarioides Grunow. Journal of applied phycology 2010;22:131-7.
[140] Vijayaraghavan K, Yun Y-S. Bacterial biosorbents and biosorption. Biotechnology advances 2008;26:266-91.
[141] Cheng R, Jiang Z, Ou S, Li Y, Xiang B. Investigation of acid black 1 adsorption onto amino-polysaccharides. Polymer bulletin 2009;62:69-77.
[142] Kousha M, Daneshvar E, Dopeikar H, Taghavi D, Bhatnagar A. Box–Behnken design optimization of Acid Black 1 dye biosorption by different brown macroalgae. Chemical Engineering Journal 2012;179:158-68.
[143] Wang S, Li H. Kinetic modelling and mechanism of dye adsorption on unburned carbon. Dyes and pigments 2007;72:308-14.
[144] Emami Moghaddam SA, Harun R, Mokhtar MN, Zakaria R. Potential of zeolite and algae in biomass immobilization. BioMed research international 2018;2018.
[145] Kalesh N, Nair S. The accumulation levels of heavy metals (Ni, Cr, Sr, & Ag) in marine algae from southwest coast of India. Toxicological & Environmental Chemistry 2005;87:135-46.
[146] Jothinayagi N, Anbazhagan C. Heavy metal monitoring of Rameswaram coast by some Sargassum species. American-Eurasian Journal of Scientific Research 2009;4:73-80.
[147] Alp MT, Şen B, Özbay Ö. Hazar Gölü'nde Mevsimsel Olarak Ortaya Çıkan Cladophora glomerata'da Bazı Ağır Metal Düzeyleri. Ekoloji 2011;20:13-7.
[148] Alp MT, Ozbay O, Sungur MA. Determination of heavy metal levels in sediment and macroalgae (Ulva sp. and Enteromorpha sp.) on the Mersin coast. Ekoloji 2012;21:47-55.
[149] Palmer C. Algae and water pollution,‖ Castle House Publications Ltd. England; 1980.
[150] Mackenthun KM. The practice of water pollution biology: US Federal Water Pollution Control Administration, Division of Technical Support; 1969.

[151] Mohan SV, Rao NC, Prasad KK, Karthikeyan J. Treatment of simulated Reactive Yellow 22 (Azo) dye effluents using Spirogyra species. Waste Management 2002;22:575-82.
[152] Daneshvar N, Ayazloo M, Khataee A, Pourhassan M. Biodegradation of the textile dye malachite green by Microalgae cosmarium sp. International Journal for Science and high Technology and Environmental Sciences 2005.
[153] Sarwa P, Vijayakumar R, Verma SK. Adsorption of acid red 66 dye from aqueous solution by green microalgae Acutodesmus obliquus strain PSV2 isolated from an industrial polluted site. Open Access Library Journal 2014;1:1-8.
[154] Hernández-Zamora M, Cristiani-Urbina E, Martínez-Jerónimo F, Perales-Vela HV, Ponce-Noyola T, del Carmen Montes-Horcasitas M, et al. Bioremoval of the azo dye Congo Red by the microalga Chlorella vulgaris. Environmental Science and Pollution Research 2015;22:10811-23.
[155] Kumar SD, Santhanam P, Nandakumar R, Anath S, Prasath BB, Devi AS, et al. Preliminary study on the dye removal efficacy of immobilized marine and freshwater microalgal beads from textile wastewater. African Journal of biotechnology 2014;13.
[156] El-Sheekh M, El-Shanshoury A, Abou-El-Souod G, Gharieb D, El Shafay S. Decolorization of dyestuffs by some species of green algae and cyanobacteria and its consortium. International Journal of Environmental Science and Technology 2021:1-12.
[157] El Nemr A, Shoaib AG, El Sikaily A, Mohamed AE-DA, Hassan AF. Evaluation of Cationic Methylene Blue Dye Removal by High Surface Area Mesoporous Activated Carbon Derived from Ulva lactuca. Environmental Processes 2021:1-22.
[158] Liu S, Ding Y, Li P, Diao K, Tan X, Lei F, et al. Adsorption of the anionic dye Congo red from aqueous solution onto natural zeolites modified with N, N-dimethyl dehydroabietylamine oxide. Chemical Engineering Journal 2014;248:135-44.
[159] Madan S, Shaw R, Tiwari S, Tiwari SK. Adsorption dynamics of Congo red dye removal using ZnO functionalized high silica zeolitic particles. Applied Surface Science 2019;487:907-17.
[160] Wang S, Peng Y. Natural zeolites as effective adsorbents in water and wastewater treatment. Chemical engineering journal 2010;156:11-24.
[161] Margeta K, Vojnović B. Development of natural zeolites for their use in water-treatment systems. Recent patents on nanotechnology 2011;5:89-99.
[162] Wiyantoko B, Rahmah N. Measurement of cation exchange capacity (CEC) on natural zeolite by percolation method. AIP Conference Proceedings: AIP Publishing LLC; 2017. p. 020021.
[163] Gadhban MY, Abdulmajed YR, Ali FD, Al-Sharify ZT. Preparation of Nano Zeolite and itsApplication in Water Treatment. IOP Conference Series: Materials Science and Engineering: IOP Publishing; 2020. p. 012054.
[164] Doula M, Dimirkou A. Use of an iron-overexchanged clinoptilolite for the removal of Cu2+ ions from heavily contaminated drinking water samples. Journal of Hazardous Materials 2008;151:738-45.
[165] Al-Moameri HH, Jaf LA, Suppes GJ. Simulation Approach to Learning Polymer Science. Journal of Chemical Education 2018;95:1554-61.
[166] Armağan B, Turan M. Equilibrium studies on the adsorption of reactive azo dyes into zeolite. Desalination 2004;170:33-9.
[167] Salima A, Benaouda B, Noureddine B, Duclaux L. Application of Ulva lactuca and Systoceira stricta algae-based activated carbons to hazardous cationic dyes removal from industrial effluents. Water research 2013;47:3375-88.
[168] Nemchi F, Bestani B, Benderdouche N, Belhakem M, de Minorval LC. Adsorption of Supranol yellow 4GL from aqueous solution onto activated carbons prepared from seawater algae. Adsorption Science & Technology 2012;30:81-95.
[169] Hameed B, Ahmad A, Aziz N. Adsorption of reactive dye on palm-oil industry waste: equilibrium, kinetic and thermodynamic studies. Desalination 2009;247:551-60.
[170] Benaïssa H, Bereksi ZS. Influence of some experimental parameters on Methylene Blue sorption kinetics from synthetique aqueous solutions using thistle stalks as a low-cost sorbent: experimental and modelling studies. studies 2012.
[171] Kazemi J, Javanbakht V. Alginate beads impregnated with magnetic Chitosan@ Zeolite nanocomposite for cationic methylene blue dye removal from aqueous solution. International journal of biological macromolecules 2020;154:1426-37.
[172] Radoor S, Karayil J, Parameswaranpillai J, Siengchin S. Removal of anionic dye Congo red from aqueous environment using polyvinyl alcohol/sodium alginate/ZSM-5 zeolite membrane. Scientific reports 2020;10:1-15.
[173] Ma Y, Qi P, Ju J, Wang Q, Hao L, Wang R, et al. Gelatin/alginate composite nanofiber membranes for effective and even adsorption of

cationic dyes. Composites Part B: Engineering 2019;162:671-7.
[174] Chen W, Ma H, Xing B. Electrospinning of multifunctional cellulose acetate membrane and its adsorption properties for ionic dyes. International Journal of Biological Macromolecules 2020.
[175] Pratiwi D, Prasetyo D, Poeloengasih C. Adsorption of methylene blue dye using marine algae Ulva lactuca. IOP Conference Series: Earth and Environmental Science: IOP Publishing; 2019. p. 012012.
[176] El Nemr A, Abdelwahab O, Khaled A, El Sikaily A. Biosorption of Direct Yellow 12 from aqueous solution using green alga Ulva lactuca. Chemistry and Ecology 2006;22:253-66.
[177] Deokar R, Sabale A. Biosorption of methylene blue and malachite green from binary solution onto Ulva Lactuca. Int J Curr Microbiol App Sci 2014;3:295-304.
[178] Tahir H, Sultan M, Jahanzeb Q. Removal of basic dye methylene blue by using bioabsorbents Ulva lactuca and Sargassum. African Journal of Biotechnology 2008;7.
[179] El Sikaily A, Khaled A, Nemr AE, Abdelwahab O. Removal of methylene blue from aqueous solution by marine green alga Ulva lactuca. Chemistry and Ecology 2006;22:149-57.
[180] Badeenezhad A, Azhdarpoor A, Bahrami S, Yousefinejad S. Removal of methylene blue dye from aqueous solutions by natural clinoptilolite and clinoptilolite modified by iron oxide nanoparticles. Molecular Simulation 2019;45:564-71.
[181] Afkhami A, Bagheri H, Madrakian T. Alumina nanoparticles grafted with functional groups as a new adsorbent in efficient removal of formaldehyde from water samples. Desalination 2011;281:151-8.
[182] Nodehi R, Shayesteh H, Kelishami AR. Enhanced adsorption of congo red using cationic surfactant functionalized zeolite particles. Microchemical Journal 2020;153:104281.
[183] Deokar R, Sabale A. Biosorption of methylene blue and malachite green from binary solution onto Ulva Lactuca. Int J Curr Microbiol Appl Sci 2014;3:295-304.
[184] Hameed B. Evaluation of papaya seeds as a novel non-conventional low-cost adsorbent for removal of methylene blue. Journal of Hazardous materials 2009;162:939-44.
[185] Mahini R, Esmaeili H, Foroutan R. Adsorption of methyl violet from aqueous solution using brown algae Padina sanctae-crucis. Turkish Journal of Biochemistry 2018;43:623-31.
[186] Abukhadra MR, Mohamed AS. Adsorption removal of safranin dye contaminants from water using various types of natural zeolite. Silicon 2019;11:1635-47.
[187] Abukhadra MR, Shaban M, Abd El Samad MA. Enhanced photocatalytic removal of Safranin-T dye under sunlight within minute time intervals using heulandite/polyaniline@ nickel oxide composite as a novel photocatalyst. Ecotoxicology and environmental safety 2018;162:261-71.
[188] Mohamed F, Abukhadra MR, Shaban M. Removal of safranin dye from water using polypyrrole nanofiber/Zn-Fe layered double hydroxide nanocomposite (Ppy NF/Zn-Fe LDH) of enhanced adsorption and photocatalytic properties. Science of the Total Environment 2018;640:352-63.
[189] Polatoğlu İ. Chemical behaviour of clinoptilolite rich natural zeolite in aqueous medium: İzmir Institute of Technology; 2005.
[190] Shaban M, Hassouna ME, Nasief FM, AbuKhadra MR. Adsorption properties of kaolinite-based nanocomposites for Fe and Mn pollutants from aqueous solutions and raw ground water: kinetics and equilibrium studies. Environmental Science and Pollution Research 2017;24:22954-66.
[191] Shaban M, Abukhadra MR, Shahien M, Ibrahim SS. Novel bentonite/zeolite-NaP composite efficiently removes methylene blue and Congo red dyes. Environmental chemistry letters 2018;16:275-80.
[192] Alver E, Metin AÜ. Anionic dye removal from aqueous solutions using modified zeolite: Adsorption kinetics and isotherm studies. Chemical Engineering Journal 2012;200:59-67.
[193] Kumar KV, Sivanesan S, Ramamurthi V. Adsorption of malachite green onto Pithophora sp., a fresh water algae: equilibrium and kinetic modelling. Process Biochemistry 2005;40:2865-72.
[194] Khataee A, Dehghan G. Optimization of biological treatment of a dye solution by macroalgae Cladophora sp. using response surface methodology. Journal of the Taiwan Institute of Chemical Engineers 2011;42:26-33.
[195] Daneshvar N, Ayazloo M, Khataee A, Pourhassan M. Biological decolorization of dye solution containing Malachite Green by microalgae Cosmarium sp. Bioresource technology 2007;98:1176-82.
[196] Kousha M, Daneshvar E, Sohrabi M, Koutahzadeh N, Khataee A. Optimization of CI Acid black 1 biosorption by Cystoseira indica and Gracilaria persica biomasses from aqueous solutions. International Biodeterioration & Biodegradation 2012;67:56-63.
[197] Özer A, Gürbüz G, Çalimli A, Körbahti BK. Biosorption of copper (II) ions on Enteromorpha prolifera: application of response surface methodology (RSM). Chemical Engineering Journal 2009;146:377-87.

[198] Shukla S, Pai RS. Adsorption of Cu (II), Ni (II) and Zn (II) on dye loaded groundnut shells and sawdust. Separation and purification Technology 2005;43:1-8.

[199] Jayaraj R, Mohan MC, Prasath P, Khan TH. Malachite green dye removal using the seaweed enteromorpha. E-journal of Chemistry 2011;8.

[200] Cengiz S, Cavas L. Removal of methylene blue by invasive marine seaweed: Caulerpa racemosa var. cylindracea. Bioresource Technology 2008;99:2357-63.

[201] Caparkaya D, Cavas L. Biosorption of Methylene Blue by a Brown Alga Cystoseira barbatula Kützing. Acta Chimica Slovenica 2008;55.

[202] El Jamal MM, Ncibi MC. Biosorption of methylene blue by chaetophora elegans algae: Kinetics, equilibrium and thermodynamic studies. Acta Chim Slov 2012;59:24-31.

[203] Indhumathi P, Syed Shabudeen P, Shoba U, Saraswathy C. The removal of chromium from aqueous solution by using green micro algae. Journal of Chemical and Pharmaceutical Research 2014;6:799-808.

[204] Liu C, Liu H, Xiong T, Xu A, Pan B, Tang K. Graphene oxide reinforced alginate/PVA double network hydrogels for efficient dye removal. Polymers 2018;10:835.

[205] El Maghraby DM. Evaluation of non-viable biomass of Laurencia papillosa for decolorization of dye waste water. African Journal of Biotechnology 2013;12.

[206] Srinivasan A, Viraraghavan T. Decolorization of dye wastewaters by biosorbents: a review. Journal of environmental management 2010;91:1915-29.

[207] Doğan M, Alkan M, Türkyilmaz A, Özdemir Y. Kinetics and mechanism of removal of methylene blue by adsorption onto perlite. Journal of hazardous materials 2004;109:141-8.

[208] Wang S, Li L, Wu H, Zhu Z. Unburned carbon as a low-cost adsorbent for treatment of methylene blue-containing wastewater. Journal of colloid and interface science 2005;292:336-43.

[209] Vadivelan V, Kumar KV. Equilibrium, kinetics, mechanism, and process design for the sorption of methylene blue onto rice husk. Journal of colloid and interface science 2005;286:90-100.

[210] Bhattacharyya KG, Sharma A. Kinetics and thermodynamics of methylene blue adsorption on neem (Azadirachta indica) leaf powder. Dyes and pigments 2005;65:51-9.

[211] Hamdaoui O. Batch study of liquid-phase adsorption of methylene blue using cedar sawdust and crushed brick. Journal of hazardous materials 2006;135:264-73.

[212] Jafari H, Mahdavinia GR, Kazemi B, Javanshir S, Alinavaz S. Basic dyes removal by adsorption process using magnetic Fucus vesiculosus (brown algae). Journal of Water and Environmental Nanotechnology 2020;5:256-69.

[213] Foroutan R, Mohammadi R, Razeghi J, Ramavandi B. Performance of algal activated carbon/Fe3O4 magnetic composite for cationic dyes removal from aqueous solutions. Algal Research 2019;40:101509.

[214] Bayat M, Javanbakht V, Esmaili J. Synthesis of zeolite/nickel ferrite/sodium alginate bionanocomposite via a co-precipitation technique for efficient removal of water-soluble methylene blue dye. International journal of biological macromolecules 2018;116:607-19.

[215] Rahimi K, Mirzaei R, Akbari A, Mirghaffari N. Preparation of nanoparticle-modified polymeric adsorbent using wastage fuzzes of mechanized carpet and its application in dye removal from aqueous solution. Journal of Cleaner Production 2018;178:373-83.

[216] Rajput S, Pittman Jr CU, Mohan D. Magnetic magnetite (Fe3O4) nanoparticle synthesis and applications for lead (Pb2+) and chromium (Cr6+) removal from water. Journal of colloid and interface science 2016;468:334-46.

[217] Liu S, Ge H, Wang C, Zou Y, Liu J. Agricultural waste/graphene oxide 3D bio-adsorbent for highly efficient removal of methylene blue from water pollution. Science of the Total Environment 2018;628:959-68.

[218] Guler UA, Ersan M, Tuncel E, Dügenci F. Mono and simultaneous removal of crystal violet and safranin dyes from aqueous solutions by HDTMA-modified Spirulina sp. Process Safety and Environmental Protection 2016;99:194-206.

[219] Mahmoud DK, Salleh MAM, Karim WAWA, Idris A, Abidin ZZ. Batch adsorption of basic dye using acid treated kenaf fibre char: equilibrium, kinetic and thermodynamic studies. Chemical Engineering Journal 2012;181:449-57.