Review Article

Azo Dye Removal Technologies

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Abstract

The implementation of such large amounts of effluent wastewater containing industrial textile coloring agents in the rest of the ecosystem to deteriorate water quality, through the generation of pollution, eutrophication, and disruption of aquatic life and constitute a serious threat to public health. Furthermore, most of the dyes and their metabolic intermediates are mutagenic and carcinogenic. The synthetic dyes are synthesized contaminants because the biosphere and is opposed to the traditional system of mineralization of organic wastewater. The azo dyes are the most important class and the largest commercial dyes and account for nearly 75% of all products of textile dyes. Physicochemical methods used to treat the wastewater of dying process have disadvantages such as excessive use of chemicals and the production of sludge disposal obvious problems. Bioremediation for the removal of dyes is gaining interest because it is obviously profitable, environmentally friendly and produces much less sludge. Therefore, for the successful implementation of a process of biological treatment of wastewater to remove azo dyes have been developed and deeply discussed in this chapter.

Keywords: Azo dye; Eutrophication; Bioremediation; Biological treatment

Introduction

Since the beginning of mankind, people have used dyes for painting and dyeing their environment, their skins and their clothes. Until the middle of the 19th century, all colorantsapplied were of natural origin. Inorganic pigments such as soot, manganese oxide, hematite and Ocher was used in living memory [1]. Paleolithic rock paintings, such as the 30,000-year-old drawings recently discovered in the caves of chauvet France provides an ancient testimony of their application. Natural organic dyes also have a timeless history of application, in particular as textile dyes. These dyes are all aromatic compounds, usually derived from plants but also from insects, mushrooms and lichens. The manufacture of synthetic dyes began in 1856, when the English chemist W. H. Perkin, in an attempt to synthesis of quinine, obtained in place of a bluish substance with excellent dyeing properties later has become a violet aniline, purple tyrant or mauveine [2]. Perkin, 18, patented his invention and set up a production line. This concept of research and development was soon to be followed by others and new dyes began to appear on the market, a highly stimulated process by the discovery of Kékulé of the molecular structure of benzene in 1865. At the beginning of 20th Century, synthetic dyes had almost completely supplanted natural dyes [3].

Classification of Dyes

Dyes are molecules with electronic systems delocalized with conjugates double bonds that contain two groups: chromophore and auxochrome. The chromophore is a group of atoms, which controls the color of the dye, and it is generally an electron withdrawing group [4]. The most important chromophores are $-C = C-$, $-C = N-$, $-C = O$, $-N = N-$, $-NO_2$ and $-NO$. The auxochrome is an electron donor substituent which can intensify the color of the chromophore by modifying the overall energy of the electronic system and providing the solubility and the adhesion of the dye to the fiber. The most important auochromes are - NH_2, $-NR_2$, $-NHR$, $-COOH$, $-SO_3H$, $-OH$ and $-OCH_3$ [5]. On the basis of the chemical structure or the chromophore, 20 to 30 different dye groups can be identified. Azo, anthraquinone, phthalocyanine and triarylmethane dyes are quantitatively the most important chromophores (Figure 1).

Dye Removal Technologies

The textile industry has been subjected to immense pressure to reduce the use of harmful substances, in particular mutagenic, carcinogenic and allergenic chemicals and textile dyes. The sanitation of textile dye sewage relies not only on the elimination of colors but also in the degradation and mineralization of dye molecules. A wide range of technologies has been developed for the removal of synthetic products dyes from water and wastewater to reduce their environmental impact [6]. These technologies includes physical methods (membrane filtration processes and sorption techniques), chemical methods (coagulation or flocculation and conventional oxidation processes), and biological methods (microbial and enzymatic degradation). The above mentioned technologies having their own merits and demerits [7]. Colors are the first pollutants to be recognized in waste water and must be removed before discharged into the watercourses. Even the presence of very small amounts of dye in water affects visual quality of water. In comparison to colorless organic compound, removal of color is more important that burdens a larger portion of biochemical oxygen demand. Methods for removing biochemical oxygen demands from most waste water are relatively well established [8]. To remove synthetic dyes from water and wastewater, a wide range of methods have been developed to reduce their environmental impact. These methods are described in the following sections.

The effluents from textile industries are the most expressive of an ecological physiological point of view [9]. To achieve satisfactory and acceptable quality levels that allow the recycling of wastewater from

textile, elimination of dyes and related compounds is very crucial. Based on the fact that azo dyes constitute the largest percentage of textile dyes, most treatment methods are based on the discoloration of azo dyes [10]. Currently, the main operational methods used in the treatment of wastewater involve physical and textiles chemical processes [11,12]. There are several factors that determine each technical and economic feasibility of dye removal technology. These includes dye, composition of waste water, dose, running costs, environmental fate and handling [13]. Each dye removal technique has its own limitations and one single process may not be sufficient to achieve complete decolorization (Table 1). To overcome this problem, dye removal strategies have a combination of different techniques. Dye removal strategies therefore essentially consist of a combination of different techniques [14].

Physico-Chemical Methods

Membrane filtration

The increased cost of water consumption and wasteful require treatment process that is integrated with the circuits of water on the ground rather than as a subsequent treatment [15]. From this point of view, membrane filtration offers a potential application. The processes that use membranes offer exciting possibilities for separation of dyestuffs and dyeing auxiliaries that reduce simultaneously hydrolysed color and biochemical oxygen demand /chemical oxygen demand of wastewater; generally used to treat effluent reactive dye bath, that could potentially reduce the volume of waste and recovery simultaneously salt [16]. On the other hand, can be separated into two or more components the flow of fluid to its molecular size. The advantages of membrane filtration are because, it is fast with low space requirement and saturation can be reused. The disadvantage with membrane filtration method has a limited lifetime before it happens fouling of the membrane and the cost is also excessive [17]. The choice of the membrane process, in the case of reverse osmosis, nanofiltration, ultra filtration or microfiltration, should be guided by the quality of the final product.

Flocculation and coagulation

These techniques are repeatedly used for the separation of suspended solids portion in the wastewater [18]. Suspended solids are remain in the wastewater and will settle out due to same type of surface charge on the particles that repel each other when they come close together. To overcome this problem, proper coagulation and flocculation techniques are used. In coagulation process, chemical coagulants bearing opposite charge, those of the suspended solids are added to the suspension that neutralizes the charges. Now the small suspended particles are capable of sticking together. Flocculation is a gentle mixing stage in which unstable particles bind with larger hairs to form a floc that can be easily removed from the suspension [19,20]. Floc size continues to build with additional collisions and interaction with added inorganic polymers (coagulant) or organic polymers. Inorganic coagulants such as aluminum and iron salts are the most commonly used coagulants. Synthetic polyelectrolytes, fly ash and clay are also used as a thickening agent [21]. However, inorganic coagulants are not very much effective to remove highly soluble dyes [22]. The greatest disadvantage of using this process is the possibility of secondary pollution [23]. Recently, some organic polymers are reported that have good coagulation property [24].

Ion exchange

Standard ion exchange systems are not extensively compatible do utilize dye of wastewater because the ion exchangers cannot catch wide range of dyes due to presence of various additives in wastewater [25]. In this technique, the waste water becomes an ion exchange resin until all available exchange sites are saturated. Both anionic and cationic dyestuffs were efficiently removed in this process. The major disadvantage of the process includes high cost of regenerating organic solvents from the ion exchanger [26].

Reverse osmosis

Reverse osmosis membranes have a retention rate of 90% or more types of ionic compounds and produces high quality permeate [27]. Degradation and elimination of chemical additives in dye waste water can be carried out in one step reverse osmosis. Reverse osmosis makes it possible to hydrolyze all mineral salts and reactive dyes and chemical aids. It should be noted that higher the concentration of dissolved salt, the more important the osmotic pressure becomes; Therefore, the greater the energy required for the separation process.

Nanofiltration

Nanofiltration has been used to treat colored wastewater from the treatment of textile dye drainage water. The adsorption step precedes nanofiltration because

This sequence reduces the concentration polarization during the filtration process, which increases process output [28]. Maintain nanofiltration membranes of low molecular weight organic compounds, divalent ions, large monovalent ions, Hydrolyzed reactive dyes and dyeing of auxiliaries. Harmful effects of high concentrations of dye and salts in dye house outflows have often been reported [29-31]. In most published studies on dye waste water, the concentration of mineral salts is not more than 20 g/l and the concentration of dye does not exceed 1.5 g/l [32]. In general, the wastewater is reconstituted with only one dye [33], and the volume studied is also low. The treatment of waste water by nanofiltration is one of the rare applications that are possible for the treatment of solutions with highly concentrated and complex solutions [34]. An important problem is the accumulation of dissolved solids, which eliminates the treated discharge outflows in water currents impossible.

Ultra filtration

Ultra filtration allows the removal of macromolecules and particles, but the elimination of pollutants such as dyes is never complete [35]. It is only between 31% and 76%. Even in the best cases, the quality of treated wastewater does not allow reuse of sensitive processes such as textile dyeing. Ultra filtration cannot be used as a pretreatment for reverse osmosis [36] or in combination with a biological reactor [37].

Microfiltration

Microfiltration is suitable for the treatment of coloring pigments containing pigment dyes [38] as well as subsequent rinsing baths. Chemicals used in the dyeing bath, which is not filtered by microfiltration, will remain in the bath. Microfiltration can also be used as a pre-treatment of nano filtration or reverse osmosis [39].

Adsorption

Processes in which ions or molecules present in one phase are

inclined to accumulate and concentrate on the surface of another phase. Physical adsorption occurs when weak intermediate bonds exist between the adsorbate and the adsorbent. Examples of such bindings are Van der Walls' interactions, hydrogen bonding and dipole-dipole interactions. In Most cases, physical adsorption is easily reversible. Chemical adsorption takes place when strong interspecies bindings are present between the adsorbate and the adsorbent by an electron exchange. Examples of such bonds are covalent and ionic bonds. Chemisorption is considered irreversible in most cases. Suzuki [40] discussed the role of adsorption in water environment processes and also evaluated the development of newer adsorbents to modernize the treatment, systems and role modeling the findings is in their development. Most adsorbents are very porous materials. Since the pores are generally very small, the internal surface is in the order size greater than the external area. Among the numerous techniques of dye removal, this technique gives the most important results because it can be used to remove others types of dyes also [40]. Waste water adsorption techniques have become more popular in recent years due to their efficiency in the removal of pollutants that are not readily biodegradable. Adsorption can produce high quality of water, while also a process that is cost-effective. Decolorize is a consequence of two mechanisms - adsorption and ion exchange and is influenced by many factors including dye/adsorbent interaction, adsorbent surface, particle size, temperature, pH and contact time. If the adsorption system is properly designed, it will produce a high quality treated waste water.

Electrolysis

Electrochemical technology is a simple process to remove a wide range of dyes and pigments. This technique is based on the use of electric power in wastewater using sacrificial iron electrode to produce iron hydroxide in solution which leads to remove soluble and insoluble acid dyes from wastewater. In addition, Fe (II) may also reduce azo dyes into aryl amines. The electrochemical system can eliminate 90% of impurities. However, the process is expensive due to high energy requirements and uncontrolled radical reactions [41].

Activated carbon

Activated carbon is a preferred adsorbent, which is widely used for wastewater treatment, which contains a variety of toner. However, the disadvantage associated with this is its high cost [42-44]. Regeneration of saturated carbon is also expensive, not simple, and leads to loss of adsorbent. Use of coal at relatively low prices the starting materials is also unfounded for most of the anti-pollution applications [45]. Various carbonaceous materials such as coal, lignite, coconuts, wood and turf is used for commercially activated carbon [46]. However, the abundance and availability of agricultural by-products makes them good activated carbon raw materials. The agricultural products of the products are renewable raw materials for the production of activated carbon since methods for reuse of waste materials are highly desirable [47]. The destruction of agricultural by-products is currently a major economic and ecological issue, conversion of by-products to adsorbents, such as activated carbon, represents a possible outlet.

Fenton

The oxidation system is based on fenton reagent - Mixture of hydrogen peroxide and iron salt has been used both for the treatment of organic matter and inorganic substances. The process is based on the formation of reactive oxidation species that are capable of effectively disintegrating contaminants on wastewaters [48]. It is accepted that both hydroxyl and ferrylic complexes occur in fenton reaction depends on the mechanism and conditions of use of which one is predominant [49]. Oxidation system can be used effectively for the destruction of non-biodegradable toxic waste water [50]. Fenton's oxidation process can stain a large range of dyes and relative to ozonization. The process is relatively cheap and leads in general in a higher chemical oxygen demand reduction [51]. Fenton oxidation is limited to that in the textile process; where waste water is generally high at pH, while the Fenton process requires low pH. At higher pH, deposition produces large amounts of waste liquor iron salts and process loss its effectiveness.

Ozonation

Ozone is a very powerful and fast oxidizing agent that can react with most of the chemicals and with simple oxidizing agent ions, such as S-2, forming oxyanions such as $SO3-2$ and $SO42$ [52]. Ozone quickly decolorizes water-soluble dyestuffs, but with non-soluble dyestuffs react much slower. In addition, the wastewater generated by the treatment of textiles generally includes other refractory components that react with ozone, which increases its demand [53-55]. Ozone degradation requires a high pH (pH> 10). In alkaline solutions, ozone reacts almost inseparably to all the present compounds [56], transforming organic compounds smaller and more biodegradable molecules [57]. Thus, ozone therapy seems logical to use biological methods to achieve complete mineralization [58]. An important limitation of ozonation is the process is a relatively high cost of ozone production process with it short half-life [59].

Advance oxidation process

An advanced oxidation process can be used to remove dyes from the waste water to produce a highly reactive radical which may be reacts a wide variety of compounds that are difficult to break down. This process includes chlorination, bleaching, photocatalytic oxidation [60].

Biological Methods

Polymers and colorants are generally difficult to biodegrade and many substances are totally unsuitable for conventional biological treatment. For textiles, in particular the emphasis is on biological treatment systems physical and chemical. biodegradation bleaching method as fungi, microbial degradation and adsorption to life dead or systems commonly applied in the treatment of industrial effluents due Many microorganisms such as bacteria, yeasts, algae and fungi are able to accumulate and degrading different pollutants [61], and all biological systems require an entry continuous effluent. Biological treatment requires a large land area and is limited by sensitivity to the diurnal variation and toxicity some chemicals, and less flexibility in design and operation. Biological treatment is unable to obtain satisfactory color removal with current conventional biodegradation processes. In addition, although many organic molecules degrade, many others are recalcitrant because of its complex chemical structure and organic synthetic to in particular, the nature of xenobiotics, the biodegradability of azo dyes is very limited.

Decolourization by prokaryotes

The ability of bacteria to metabolize azo dyes has been investigated by a number of research groups [62]. Under aerobic conditions azo dyes are not readily metabolized, though the ability of bacteria with specialized reductive enzymes to aerobically degrade certain azo compounds [63]. In contrast, many bacteria are anaerobic which reduces azo dyes by non-specific, soluble, cytoplasmic reductase activity. Anaerobic reduction of degrades azo dyes may be convert into aromatic amines [64], which may be toxic, mutagenic, and possibly carcinogenic for mammals [65]. Therefore, in order to achieve complete degradation of azo dyes or azo compounds this involves the aerobic biodegradation of the produced material [66]. In phthalocyanine colors, reversible reduction and discoloration are present under anaerobic conditions [67].

Decolorization by fungi

The most studied fungi with respect to the degradation of dyes are ligninolytic fungi that produce enzymes such as lignin peroxidase, manganese peroxidase and laccase [68]. There are much literature on the potential use of these fungi to oxidize phenolic, non-phenolic, soluble and insoluble dyes [69]. Species of Pleurotus ostreatus, Schizophyllum, Sclerotium rolfsii and Neurospora crassa appeared to increase up to 25% the degree of discoloration of particular textile dyes like triaryl methane, anthraquinone and indigoide using enzymatic preparations [70]. In contrast, manganese peroxidise has been reported as the main enzyme involved in color discoloration by Phanerochaete Chrysosporium [71] and lignin peroxidase for Bjerkandera adusta [72]. Some non-white-rot mushrooms that can successfully discolor dyes also reported by the researchers [73].

Decolorization by yeast

In the literature, the ability to degrade azo dyes by yeasts has been described only in some respects. The first two reports describes the use of Ascomycete and Candida zeylanoides yeast isolated from contaminated soils to reduce model azo dyes [74]. Characterization of enzymatic activity is described in other studies with yeast Issatchenkia occidentalis [75], and the enzymatic system involved in a work with Saccharomyces cerevisiae [76].

Decolorization by algae

Only few algae are reported for the degradation of dyes such as Chlorella, Oscillateria [77] and Spirogyra [78] (Figure 2). Jinqi and Houtian [77] also indicate that certain azo compounds tested could be used as sources of carbon and nitrogen. This could mean that algae can play an important role in the elimination of azo dyes and aromatic amines in stabilization ponds [78].

Conclusion

This opinion makes a simple comparison between different physicochemical methods namely photocatalysis, electrochemical adsorption, hydrolysis and biological methods; also discussed the advantages and disadvantages of these methods involved in azo dye decolorization. The main disadvantages of physical methods such as adsorption, ion exchange and membrane filtration were simply transferred dye molecule to another phase rather than destroy them and are only effective when the volume of effluent is small. The main disadvantage of chemical methods such as chemical oxidation, electrochemical degradation and ozonation were the requirements of

an effective pretreatment sludge production. Biological degradation, bioaccumulation and biosorption were the three most important technologies used in biological dye removal process. They have a good potential to replace conventional methods for the treatment of dye carrier of industrial wastewater. Biological processes can be applied in situ at the infected site, these were usually environmental friendly, that is, no secondary pollution and they were cost effective. These were the principle benefits of biological technologies for treatment of industrial waste. Hence, in recent years, the focus of the research is strongly focused biological methods for the treatment of waste water. The downside of the degradation process is its low degradation efficiency for some dyes and practical difficulties in continuous method. The major disadvantage of bioaccumulation process is using living organism that is not advisable for the continuous treatment of highly toxic wastewater. This problem can be overcome with biosorption by using dead biomass. In this context, the literatures suggested that biosorption is an inoffensive and a cost-effective method; and also does not produce any secondary pollutants.

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