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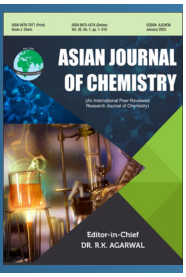

REVIEW

Removal of Colours in Textile Wastewater by Electrochemical Methods: A Review

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Dye wastewater from industries poses significant health hazards to the environment, so it is important to limit its discharge into receiving waters. This overview discusses several feasible, low-cost treatment methods, which includes biological, chemical and physical approaches. Adsorption and membrane filtration are the two common and widely acceptable physical methods, whereas chemical (or widely known as oxidative methods include Fenton treatment, ozone treatment, H_2O_2 UV irradiation, hydrogen peroxide, NaOCl, ion-exchange, electrocoagulation, reverse process, nanofiltration, *etc.*). Biodegradation can occur either aerobically or anaerobically, and both of these are examples of the biological techniques. When it comes to the treatment of wastewater effluents from the textile industry, there are primarily two areas of concern: selecting the appropriate technology and developing an appropriate treatment strategy. However, The treatment of dye solutions typically depends heavily on electrochemical processes.

Keywords: Wastewater from textiles, Treatment methods, Chemicals, Dyes, Pollution.

INTRODUCTION

Water, the priceless natural resource that is necessary for the survival of every living thing on earth, is essential to all forms of life. Rapid population development, accompanied with increased industrialization and urbanisation, is now responsible for both water shortages and water pollution. Roughly 7×10^5 – 1×10^6 tons of dyes generated each year, however, only 10-20% of the total dyes are generally utilized in the world. It is believed that over 100,000 different types of commercial dyes can be found in wastewater generated all over the world [1]. In the textile industry, sewage is discharged directly into the water bodies that adverse affect the environment [2]. A wide range of industries use synthetic dyes, including textile staining, paper printing, colour photography, pharmaceutical, paper, pulp, cosmetic and other industries [3]. Pollutants in wastewater reduce the amount of light reaching aquatic life, which in turn reduces photosynthesis, increases the need for oxygen and has a negative impact on the ecosystem [4].

Regeneration, reuse and recycling are the three main approaches to wastewater's waste reduction. As a result of this process, wastewater is turned into operational water that can be reused for a variety of purposes. Partial treatment reduces pollutants in wastewater, allowing it to be reused for a variety of industrial and domestic uses after regeneration. In order to comply with the discharge standards, an efficient method to treat wastewater containing dye is urgently needed [5]. Advanced oxidation processes (AOPs) and electrochemical technologies have been developed for the treatment of contaminants in drinking water and industrial effluents [6].

In electrocoagulation, iron or aluminum anodes are electrodissolved within wastewater to form metallic hydroxide flocs. Despite its limited success, this method has been practiced for most of the 20th century. However, due to the restrictions in regulating the effluent wastewater, there has been a rise in interest in electro-coagulation in recent years. In developed countries, this technology has become increasingly popular for treating industrial wastewater in the past decade [7].

A modification of electrofloatation and electrocoagulation involves the production of coagulating agents *in situ* at the electrodes (iron and aluminum are the most commonly used electrodes). At anodes, precipitates or adsorbed hydroxides are formed when the pollutant is converted to amorphous precipitates. Electroflocculation is another name for this method, in this proceess, anodes of mild steel/aluminum and cathodes of stainless steel are used at a range of current densities (0.25-1.0 A/dm^2 [8].

Mechanism of electrocoagulation: A common characteristic of iron electrodes is the transfer of large numbers of Fe$^+$ ions into solutions, resulting in the production of more sludge. On account of its low cost, iron electrode makes a significant contribution to efficiency when compared with other electrodes. It is more efficient to remove dye with iron electrode pairs by electrocoagulation than with copper electrode pairs. The Fe(OH)$_n$ is produced by two mechanisms [9]:

Anode:

$$4Fe\ (s) \rightarrow 4Fe^{2+}\ (aq) + 8e^- \qquad (1)$$

$$4Fe^{2+}(aq) + 10H_2O\ (l) + O_2(g) \rightarrow 4Fe(OH)_3(s) + 8H^+\ (aq) \qquad (2)$$

Cathode:

$$8H^+\ (aq) + 8e^- \rightarrow 4H_2\ (g) \qquad (3)$$

Overall:

$$4Fe\ (s) + 10H_2O\ (l) + O_2\ (g) \rightarrow 4Fe(OH)\ (s) + 4H_2\ (g) \qquad (4)$$

Insoluble iron hydroxides can absorb contaminants by surface complexation or electrostatic attraction and then flush them away. Pre-hydrolysis of Fe^{3+} cations during wastewater treatment causes the production of reactive clusters.

In wastewater treatment, electrochemical oxidation has long been a topic of interest. The development of new anodes for the degradation of organic molecules has also attracted a great attention [10-23]. Several electrodes *e.g.*, graphite electrodes [24], iron electrodes [25], PbO$_2$ electrodes [26], noble metal electrodes [27,28], dimensionally stable anodes (DSA) [29-31] and boron-doped diamond electrodes (BDD) [32-34] are among the electrodes commonly used in electrochemical oxidation. These two methods are considered to be among the best among those reported in recent years. In indirect organic pollutant degradation, dimensionally stable anodes (DSA) is increasingly used due to its high efficiency at oxidizing chloride ions [31]. Dye mineralization would be limited by active chlorine (OCl, HOCl and Cl$_2$). Besides hydroxyl radicals, boron-doped diamond (BDD) was also exposed to hydrogen peroxide radicals through water oxidation. Boron-doped diamond (BDD) electrodes are effective for removing colour and mineralizing thoroughly. Using methyl red as a catalyst, Cerisola & Panizza [35] compared the catalytic activities of four electrode materials (TiRuSnO$_2$, BDD, PbO$_2$ and Pt). Bulk electrolysis of methyl red showed complete colour and COD removal with BDD and PbO$_2$, but only partial oxidation with Pt and TiRuSnO$_2$. According to Chen *et al*. [36], Ti/BDD electrodes oxidized pollutants better than Ti/SnO$_2$–Sb$_2$O$_5$.

Mechanism of electrooxidation

Direct oxidation: There are two stages to direct oxidation of pollutants (i) diffusion to the anode surface from the bulk solution and (ii) oxidation at the anode surface. Electrochemical oxidation relies on a correlation between mass transfer and electron transfer at the electrode surface to be effective. Electron transfer rate is determined by current density and electrode activity. It is generally accepted that organic substances are oxidized by anodic current in one of two ways [37], as shown below:

(A) Electrochemical conversion: Eqn. 1 presents the partial oxidation of organic substances (R). However, to degrade the oxidized substrates completely, a subsequent treatment is necessary

$$R \rightarrow RO + e^- \qquad (1)$$

(B) Electrochemical incineration (combustion): Organic substances provide the basis for the formation of water, carbon dioxide and other inorganic constituents.

$$R \rightarrow CO_2 + H_2O + Salts + e^- \qquad (2)$$

Indirect oxidation: Organic compounds in bulk solutions are destroyed by indirect electrochemical oxidation as a result of electrogeneration of a strong oxidizing agent at the anode surface. Chloride is oxidized at the anode to chlorine, which is widely used as an electrochemical oxidant. Oxidation of inorganic and organic matters is possible through indirect oxidation, which uses chlorine, hypochlorite, hydrogen peroxide and peroxodisulfuric acid [38,39]. Electrochemical oxidation of wastewater removes impurities primarily through indirect oxidation, using chlorine/hypochlorite produced by anodic oxidation of chlorine. An indirect oxidation reaction involving chlorine and hypochlorite is presented in eqns. 3-9:

Anodic reactions:

$$2Cl^- \rightarrow Cl_2 + 2e^- \qquad (3)$$

$$6HOCl + 3H_2O \rightarrow 2ClO_3^- + 4Cl^- + 12H^+ + 1.5O_2 + 6e^- \qquad (4)$$

$$2H_2O \rightarrow O_2 + 4H^+ + 4e^- \qquad (5)$$

Bulk reactions:

$$Cl_2 + H_2O \rightarrow HOCl + H^+ + Cl^- \qquad (6)$$

$$HOCl \rightarrow H^+ + OCl^- \qquad (7)$$

Cathodic reactions:

$$2H_2O + 2e^- \rightarrow 2OH^- + H^+ \qquad (8)$$

$$OCl^- + H_2O + 2e^- \rightarrow Cl^- + 2OH^- \qquad (9)$$

Hypochlorite is a strong oxidizing agent that can oxidize organic compounds in bulk solutions (eqns. 6 and 7) [39]. Metal ions, however, can result in the more toxic effluent than its initial state after treatment. Due to this, a system of this kind requires a separation step to recover the metallic species [40], resulting in an unfavourable complicated treatment procedure.

Role of nanomaterials in colour removal of wastewater: One of the most widely discharged contaminants, organic dyes, can be difficult to remove using traditional wastewater treatment technologies. Over the past few decades, nanotechnology has advanced at a rapid pace, opening up new possibilities for the creation of low-cost, high-efficiency techniques of treating wastewater polluted with dyes [41]. Due to their large surface area, modified surface properties, unique electron conduction properties, *etc.*, nanomaterials have excellent performance in dye-contaminated wastewater treatment. Among the most notable is the 875.0 mg/g adsorption capacity of agar-modified monometallic/bimetallic nanoparticles for methylene blue, which is several times higher than that of conventional adsorbents [42]. Most promising nanomaterials for removing dyes from water include carbonaceous nanomaterials, nanosized TiO_2 and graphitic carbon nitride (γ-C_3N_4) [43]. However, their engineering application is limited by some challenges, such as high cost and poor separation performance. The modification methods for improving the effectiveness of nanomaterials are highlighted. Finally, the current knowledge gaps of developing nanomaterials on the environmental application are discussed and the possible further research direction is proposed.

TiO_2: As a catalyst, TiO_2 has aroused great interest due to its potential on water splitting [44,45]. As a result of its high photocatalytic activity, resistance to photocorrosion, low toxicity and low cost, TiO_2 is one of the most studied and used photocatalysts at present [46]. Using TiO_2 nanoparticles for photocatalytic degradation of naphthol blue black dye has been reported [47]. Numerous researchers subsequently investigated the effects of TiO_2 on dye degradation under UV irradiation [48,49]. The surface properties of TiO_2 and the molecular structure of dyes influence dye removal efficiency. For example, it has been compared the photocatalytic degradation of eight cationic, five anionic and three solvent dyes by a combustion-synthesized nano-TiO_2 and a commercial nano-TiO_2 (P25) under UV light, where combustion-synthesized nano-TiO_2 showed higher photocatalytic degradation efficiency for all the anionic dyes, while P25 was better at degrading most of the cationic dyes [50]. Decolorization of solvent dyes, however, was dependent on adsorption. However, TiO_2 is still limited in its efficiency as a model photocatalyst due to its low visible light activity and fast electron-hole pair recombination rate. According to the time-resolved spectroscopic studies [51,52], 95% of the electron-hole pairs were recombined, which may account for TiO_2's low quantum efficiency. Meanwhile, the wide band gap (3.2 eV) makes the TiO_2 only active under UV region. It has been attempted to overcome these limitations by promoting charge separation or lowering the band gap energy of TiO_2, including non-metal doping, transition metal doping, metal doping/deposition, dye sensitization, heterojunction, morphology control, *etc.* [53-55].

Addition of S, N, C, *etc.* to TiO_2 may extend the visible light activity by continuously modulating the band gap [56,57] synthesized the S and C-doped TiO_2 and evaluated their effectiveness by photocatalytic degrading reactive black 5, methylene blue and methyl orange. As a result of the S and C doping on the catalysts, the dye removal time was reduced from 1-4 h to less than 5-20 min. In addition to improving visible light absorption, N doping on TiO_2 retards electron-hole recombination and optimizes electronic properties [58]. Using metal doping/deposition, *e.g.*, Au, Pt, Ag and Pd, facilitates electron-hole separation and red shifts the optical absorbance. According to the results of comparing the TiO_2 that is deposited with Pt, Au and Pd by the photooxidation of acid green 16 under UV irradiation, all of the metal-deposited TiO_2 displays higher photocatalytic activity and in the order of Pt, Au and Pd [59]. Metal-doped TiO_2 had an absorption threshold that extended into the visible range as determined by UV-Visible diffuse reflectance measurements. Deposition of metal could also induce the formation of Ti^{3+} defect sites, which can adsorb and release oxygen when exposed to light. It should be noted, however, that there is an optimal metal dosage which maximizes photocatalysis efficiency and minimizes surface active site blockage.

ZnO: As a photocatalyst, zinc oxide (ZnO) is utilized to degrade various contaminants in aqueous media due to its unique qualities as an n-type semiconductor, such as its 3.3 eV direct band gap and 60 meV binding energy [60]. The unique characteristics of ZnO nanoparticles and their conjunction with modern treatment approaches facilitate ample openings to develop remediation of polluted waters [61,62]. Photoelectrocatalytic (PEC) degradation of numerous organic contaminants has attracted great attention with ZnO nanoparticles immobilized on conductive substrates [63]. Synthetic methods can manipulate the microstructures of ZnO, such as its morphology, crystal size, orientation, *etc.* to modify its properties [64]. There is a higher photocatalytic activity in nanosized ZnO synthesized with different hierarchical structures, such as flower-like, sea-urchin-shaped, dand-elion like, compared to monomorphological ZnO [65]. The ZnO nanoparticles prepared by codeposition and sol-gel methods have also shown high activity in degrading organic dyes under UV light. SiO_2 gave smaller particles and higher activity than sol-gel-prepared ZnO [60]. A larger surface area can contribute to the improved activity of ZnO nanomaterials, which in turn can improve their quantum efficiency by extending their spectrum absorbance and delaying excited electronhole recombination, similar to other pristine semiconductors. In order to suppress unwanted charge carrier recombination, metals or non-metals are doped or coupled with other semiconductors [66]. For instance, the NiO-ZnO nanocomposites showed higher photocatalytic decolorization efficiency of organic dyes under UV-visible light than pristine ZnO and are comparable to Degussa TiO_2 (P25) [66]. The incorporation of high electronical conductive compounds, *i.e.* graphene with ZnO nanoparticles can facilitate the transport of photoexcited electrons and thus promote the photocatalytic activity [67]. Meanwhile, the excited dyes can inject electrons to graphene due to the higher redox potential of graphene. Therefore, the electron-accepting properties of graphene can enhance the photocatalytic performance of the graphene-modified ZnO [68,69]. Meanwhile, the photocatalytic activity of ZnO could be improved by immobilization of ZnO nanoparticles onto siliceous materials [70].

Ag-based photocatalysts: The Ag series photocatalysts have aroused great attention due to the high visible light activity and potential application in environmental remediation. Their photocatalytic performance could be improved by morphology and facet-controlled processes, coupling with other functional materials [71]. For example, when the Ag_2O, Ag_2CO_3 and Ag_2O/Ag_2CO_3 core-shell nanoparticles with TiO_2 tested on methyl blue degradation under visible light, all these three materials showed much higher activity than TiO_2 and their precursors [72]. The hierarchical heteronanostructures of Ag-based nanomaterials could facilitate their separation of excited electron-hole pairs [73]. And the ratio of Ag salts and the functional materials are key parameters that control the photocatalytic activity. A type of Ag/AgCl core-shell nanowire was synthesized by hetero epitaxial growth for photocatalytic degrading methyl orange under visible light. The fast photodegradation was achieved at the optimum Ag/AgCl ratio of 8/92 for the core-shell nanowires, which could reach the complete methyl orange removal in 8 min under visible light [56,74]. The researches on the Ag-based photocatalysts have made considerable advances; however, further developments are needed and they are still far from the practical applications. The detailed mechanism on the photocatalytic degradation of organics is unclear and the cost of Ag-based photocatalysts is high. Moreover, due to the relatively low stability of Ag salt under irradiation, the loss of material mass and photocatalytic activity after reusing still need to be well evaluated.

Conclusion

The electrochemical method provides control conditions for the treatment of wastewater containing heavy metals, which is relatively simple and convenient to remove particulates, organic matter and inorganic matter. Although electrochemical technologies are still in their infancy in several countries, however, significant progress has been made in the last 20 years. The most effective method of waste and byproduct collection is electrocoagulation. Performance during degradation was more effective and depends on how much energy is used to remove the pollutants. Increased efficiency, economical feasible, easy operating and control conditions are in electrochemical methods. Due to which, always given preference above the other wastewater treatment technologies.

CONFLICT OF INTEREST

The authors declare that there is no conflict of interests regarding the publication of this article.

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