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Removal of Acid Orange 7 Dye from Wastewater: Review

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Abstract

The most recent removal methods for the removal of Acid Orange 7 (AO7) dye from wastewater are complied. Microbial bio-degradation, chemical decomposition by oxidation, photo-degradation and adsorption by various adsorbents are various methods used in removal of AO7. The advantages and disadvantages of the various methods are discussed and there efficiencies are compared.

Keywords: Acid orange 7; Wastewater; Adsorption; Oxidation; Biodegradation

Introduction

Synthetic dyes are extensively used in many industrial fields, such as dyestuffs, textile, paper and plastics. It is estimated that there are around 100,000 commercially available dyes with over 7×105 tons of dyestuff are produced annually [1-4]. Synthetic dyes exhibit considerable structural diversity, and, on an industrial scale, they can be classified into azo, anthraquinone, sulfur, indigo, triphenylmethyl (trityl), and phthalocyanine derivatives [5,6]. The majority of synthetic dyes currently used in the industry are azo dyes and their derivatives because they are highly stable during washing, lightfast and not susceptible to degradation under natural conditions [7,8]. Azo dyes contain one or more azo bonds (-N=N-) as chromophore groups linked to aromatic structures with functional groups such as –OH, and $-SO_3H$ [8]. Even small amounts of dyes (of the order of a few ppm) are undesirable as they colorize water, make it look unaesthetic, and disturb life processes in it. Most dyes are non-biodegradable, hinder sunlight penetration and inhibit the photosynthesis process and increase the chemical and the biological demand for oxygen. Therefore, water ecosystems are disturbed [9,10].

Anionic azo dyes, in the presence of a sulfonate (SO^{3-}) groups, will become strongly water solubilizing substituents. In aqueous solution, the negatively charged anionic azo dyes exist over a broad pH range. Anionic monoazo dyes and their metal salts are widely used on dyeing papers and leather, or as pigments [5]. Among monoazo dyes acid orange 7 [p-(2-hydroxy-1-napththylazo) benzene sulfonic acid] is the most commonly used anionic dye because it is water soluble. Acid orange 7 (AO7) is also well known, inexpensive and dyes rather quickly in weak acidic solution. Because of these properties, it is extensively used for dying a variety of materials such as nylon, aluminum, detergents, cosmetics, wool and silk [5,7,11].

Like most other azo dyes, acid orange 7 tends to be disposed in industrial waste water and possess health threats to humans [9]. It is highly toxic, and its ingestion can cause eye, skin, mucous membrane, and upper respiratory tract irritations; severe headaches; dizziness; nausea; and loss of bone marrow leading to anemia [12]. Its consumption can also prove fatal, as it is carcinogenic in nature and can lead to tumors. AO7 tends to withdraw electrons from the azo group, leading to a deficiency of electrons, thereby reducing it to carcinogenic amino compounds. The reduction of AO7 produces 1-amino-2-naphthol, which has been reported to induce bladder tumors [5,7,11]. Inside the human body AO7 can also easily undergo enzymatic breakdown, along with reduction, and forms aromatic amines, which, upon exposure, can cause methemoglobinemia. The intermediate amines thus formed also tend to oxidize the heme iron of hemoglobin from Fe (II) to Fe (III) and block oxygen binding, resulting in some characteristic symptoms such as cyanosis of lip and nose, weakness, and dizziness [12,13].

Purification of waste water containing AO7 dyes is becoming a more important goal to avoid the potential threats to the environment. The discharge of waste water contaminated by AO7 azo dyes is strictly regulated in many countries for aesthetic reasons and because of breakdown products which are toxic and mutagenic [7]. Because of the toxicity and carcinogenic nature of the AO7, researchers have tried biological, chemical and/or physical decolorization techniques (oxidation, coagulation, flotation. and sorption, respectively) to remove this colored effluent [6,14,15]. Considering the effects of AO7, removal of AO7 has been attempted using different biodegradation methods. Biological treatment is the most economical, valuable, and simple method of decolorization of azo dyes when compared with other physical and chemical procedures. Even though it is an economically attractive process, drawbacks exist. Besides being a slow process, the method also needs optimally favorable conditions, certain nutrition requirements, and large area for AO7 to decolorize. This leads to less flexibility in design and operation of biological decolorization methods. Anaerobic decolorization of AO7 uses a pure bacterial or mixed microbial consortium which leads to increase the toxicity of AO7 by nearly 100-fold, (AO7 EC50=15.7 mg/L and AO7 with *E. faecalis* EC50=0.2 mg/L) due to the production of 1-amino-2-naphthol (EC50 0.1 mg/L) [16]. Co–metabolism in a Microbial Fuel Cell (MFC) (*S. oneidensis*) utilized rapid AO7 decolorization kinetics at low to moderate AO7 concentrations. The decolourization kinetics constants (k) observed in the study above is comparable or better in comparison to that of other studies utilizing pure or mixed microbial cultures (Table 1).

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MFC (*S. oneidensis*) can give 98% decolorization on AO7 within 30 h, and during decolorization, it gives 1-amino-2-naphthol as a metabolite, which is a toxic product for humans [18,21]. The toxicity of the decolorized effluents increased when the initial AO7 concentration was raised. The half maximal inhibitory concentration (IC50) decreased from 107.1 mg COD L^{-1} at 70 mg L^{-1} initial concentration to 51.3 mg COD L^{-1} at 350 mg L^{-1} initial AO7 concentration. This result indicates increased acute toxicity at high AO7 decolonization metabolite concentrations [19]. Constructed wetland is nowadays a well-accepted eco-friendly method to treat the waste water systems. The aerated wetlands reactors remove organic matter and aromatic amines more effectively than a non-aerated one within 3 to 6 days at 100 mg L^{-1} initial AO7 concentration. Aerated wetlands processes require long times (around 3–6 days) yielding complete decolorization and mineralization of AO7 [22].

To overcome these problems, the use of Advanced Oxidation Processes (AOPs) with hydrogen peroxide (H_2O_2), ozone (O_3) and titanium dioxide (TiO_2) have been widely proposed to treat AO7 waste water systems. The AO7 was successfully removed on TiO_2 in visible light in the presence of oxygen. AOPs are a group of processes based on the generation of highly reactive radicals, especially by hydroxyl radicals, which are extremely active and nonselective oxidants. Such intermediates are able to oxidize a wide range of compounds that are otherwise difficult to degrade [23,24]. AOPs methods are often expensive are also generate powerful oxidizing agents (hydroxyl radicals) to produce concentrated sludges, thereby creating secondary pollution problems [14]. Table 2 shows the decolonization capacity of AO7 in various oxidative processes and disadvantages of each system for comparison. Decolorization results of AO7 using oxidation methods indicate it is mostly applicable to less concentrated effluents. The major disadvantage of using the oxidation method is the possibility of formation of toxic byproducts even from biodegradable substances [25].

In accordance with data from the literature, liquid–phase adsorption is the most popular technique for the removal of pollutants from waste water systems [5,32-34]. If the sorbent is inexpensive and does not require additional pre-treatment steps before application, it can be a cost effective method for the decolorization of an azo dye [14]. Among all the adsorbent materials proposed, activated carbon is the most popular and most effective adsorbent for the removal of pollutants because, as a porous material, it has many advantageous physical properties. Such properties include high surface area, high capacity and high rate of adsorption. The process has fast kinetics and yields high–quality treated effluent [35,36]. However, activated carbon has several disadvantages. It is quite expensive (recently the market price of activated carbon of industrial grade is about US $ 20-22.00/kg [36], and performance is dependent on the type of carbon used. Its dye removal performance requires a complexing agent.

According to the type of complexing agent used, the quality of purification can be changed. The better the complexing agent, the higher the efficiency, which makes the cost higher. Thus, wastewater treatment is a cost challenging problem especially in developing countries [14,35,37,38]. In order to decrease the cost of treatment, attempts have been made to find inexpensive alternatives to remove AO7 such as activated carbon obtained from organic wastes, for example, from fruit seeds and peels. This type of carbon sorbent can be applied as an affective and cheap substituent for commercial activated carbon [10]. Table 3 shows the adsorption capacity of AO7 in different systems. Results indicate that activated carbon obtained from organic wastes is unable to give as high an adsorption as activated carbon.

Because conventional adsorbents are expensive, many factories in the textile industry try to use non-conventional low cost adsorbents to remove textile dyes. Some of the reported adsorbents include clay materials (bentonite, kaolinite), zeolites, agricultural waste (rice husk, coconut shell) siliceous material (silica beads, perlite) and biosorbents

AO7 concentration (mg L^{-1})	Removal rate (mg L^{-1} d^{-1}) (dye conc.)	Decolorization kinetic constant, k (h^{-1}) (dye conc.)	Culture	Reactor type
30–300	NG*	0.1 (60 mg L^{-1}) 0.08 (300 mg L^{-1})	Methanogenic and mixed anaerobic cultures [17]	Anaerobic batch reactor
60-300	300	0.066 (300 mg L^{-1})	Anaerobic sludge from pulp and paper wastewater treatment plant [18]	Up flow blanket (UASB) anaerobic sludge
35-350	254.6 (35 mg L^{-1}) 551 (195 mg L^{-1}) 290 (350 mg L^{-1})	0.709 (35 mg L^{-1}) 0.352 (195 mg L^{-1}) 0.05 (350 mg L^{-1})	*Shewanella oneidensis* [19]	Dual chamber MFC
10-200	NG*	0.082 (10 mg L^{-1}) 0.04 (100 mg L^{-1}) 0.020 (200 mg L^{-1})	*Alcaligens faecalis* and *Rhodococcus erythropoli* [20]	Erlenmeyer flasks

Table 1: Comparison of AO7 removal rates, first-order kinetic constants of Decolorization, different inoculum and reactor types reported in other studies. NG* = Not Given.

AOPs	Adsorption capacity of AO7	Disadvantages
Photosensitization on TiO_2 particles [26]	0.10 mmol AO7/g TiO_2 (35 mg/g)	Can decolorize efficiently solutions. Produces 1,2-naphthoquinone and phthalic acid during the course of degradation, 1,2-naphthoquinone reacts with ocular tissues and is responsible for production of cataract and damage to the retina [27]
Solar photoelectrocatalytic degradation using TiO_2 as photoanode (atmospheric plasma spray used to coat TiO_2) [8]. Adsorption onto mesoporous TiO_2 nanotude and photocatalytic degradation [28]	15 mg/L AO7, pH 7.0 and anode=1.0 mA cm^{-2} 130 mg AO7/g TiO2 (Co=600 mg/L)	Can decolorize efficiently solutions with low contents of AO7 azo dye [8]. With higher anodic current density it produces large amount of OH radicals [8]
Activated carbon/Fe Fenton's reagent (H_2O_2+Fe^{2+}) [29]	98% AO7 removal (Co=0.1 mM, pH 3, [Fe^{2+}]=0.081 mg/L, T=30°C and activated carbon impregnated with iron=0.1 g/L)	Increase of the iron concentration leads to increase leaching of the iron to the solution [29]. Expensive process due to the usage of activated carbon H_2O_2 has low oxidation power (1.78 v) [30].
Electrochemically generated Fenton's reagent [31]	0.1 mM AO7, pH 3, [Fe^{3+}]=0.1 mM and I=300 mA	Formation of large number of aromatic intermediates

Table 2: Decolorization capacity of AO7 azo dye in various oxidation methods.

Adsorbent	Capacity (mg g^{-1}) or removal (%)	Concentration range mg/L
Guava seed (*Psidium guajava* L.) [39]	100%	Co=30
Bottom ash [11]	68%	Co=35
De-oiled soya [11]	58%	Co=35
Brown coal fly ash [40]	82.8 mg/g	17-70
Chemically treated wood shavings [41]	179.7 mg/g	Co=350
Oxihumolite [42]	50 mg/g	30-1400
Powdered activated carbon [43]	92%–96%	Co=150

Table 3: Adsorption capacity of AO7 on powdered activated carbon and activated carbon obtained from organic waste.

Adsorbent	Capacity (mg g^{-1}) or removal (%)	Concentration range (mg L^{-1})	Adsorption decreased when pH increased in the range
Chitosan [48]	1120	C0=350	4<pH<7
Chitosan [49]	1940	100-4000	3<pH<8

Table 4: Adsorption capacities of AO7 on cross-linked chitosan.

(chitosan-based material, peat). Among these non–conventional low cost adsorbents, chitosan is produced by alkaline N-deacetylation of chitin, which is widely found in the exoskeleton of shellfish and crustaceans. It was estimated that chitosan could be produced from fish and crustaceans at a market price of US $ 15.43/kg [36]. It's a cationic polyelectrolyte due to the presence of a large amount of primary amino groups [44]. In acidic solution, the amino groups of chitosan are easily protonated. Therefore, chitosan is able to form strong electrostatic interactions between anionic dyes, and this interaction helps to remove anionic dyes [44-48]. Table 4 gives an indication of the adsorption capacities of AO7 on cross-linked chitosan.

Although it is an environmental friendly, low cost adsorbent, chitosan has some drawbacks. Its absorption capacity depends on the origin of the polysaccharide and the degree of N–acetylation, and it is non–porous and pH dependent [35]. According to the Table 4, chitosan is able to form higher adsorption capacities with the pH less than 4. In industries, acetic acid is often used as a stimulator in the dying process, in which the pH of the dye solution is normally adjusted to 3–4. However, chitosan formed gels below pH 5.5 and could not be evaluated. Therefore, acid effluent could severely limit the use of chitosan as an adsorbent in removing dyes due to chitosan's dissolution tendency in the acid effluent [49].

The use of adsorption techniques offers potential advantages in the treatment of AO7 containing effluents. As discussed in previous paragraphs, there are many low cost adsorbents being developed to replace activated carbon. Most of non-conventional adsorbents are unable to give higher adsorption capacity on AO7, except chitosan. Consequently, some researchers used synthetic anion exchanges to get higher decolorization rate. Purification of AO7 from waste water has been studied using three different anion exchanger's resins as give on Table 5 [10]. These results show that in the presence of quaternary ammonium ions, AO7 is able to obtain higher adsorption and desorption capacities.

Conclusion

This review article presented current publications in the field of the removal of AO7 azo dye from wastewater with help of adsorption, oxidation and biodegradation. All the decolorization methods discussed on removal of AO7 have advantages and drawbacks.

AO7 colored wastewater treatment using biodegradation is the most economically valuable choices compared with other treatment techniques, due to the less running cost. However, biological treatment required long processing time to yield complete removal of AO7 colour and mineralization of the byproducts. Another drawback of biodegradation, with increasing AO7 initial concentration more toxic products are tend to make and finial toxicity level increase.

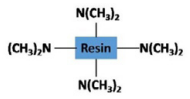
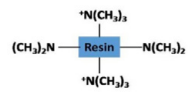
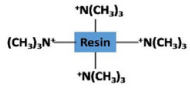
Anion exchange resin	Capacity (mg g^{-1})	Desorption %
Weakly basic 	769.5	95-100% KSCN with methanol
Intermediate basic 	979	~88% KSCN with methanol
Strongly basic 	1004.4	90% with 1 M HCl

Table 5: Different amino functionalized anionic resins [10].

Wide ranges of natural adsorbents were used on aqueous wastewater systems for the removal of AO7, due to there are inexpensive, locally available and effective material could be used in place activated carbon. Among non–conventional and conventional adsorbents, chitosan and anion exchange with quaternary ammonium ions, were able to show higher adsorption capacities on removal of AO7. Although much has been accomplished in the area of removal of AO7, much work is necessary on improving the adsorption capacity of AO7 and demonstrates the use of treatment methods on large scale.

Among adsorption and biological methods, advance oxidation techniques are still a costly-comparative substitute for the treatment of AO7 wastewaters and wildly able to practice by the small and large scale industries. Main disadvantage of oxidation treatment is the formation of toxic products, during degradation process.

However, until to date the applicability of adsorption, oxidation and biodegradation methods for the removal of AO7 is very limited on industrial level. More and more studies required to estimate their application for colour removal of AO7 particularly, their performance at higher concentration of AO7, pH, organic matter and heavy metal etc.

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