Applied Water Science (2022) 12:173
https://doi.org/10.1007/s13201-022-01695-3

REVIEW ARTICLE

Comparison of available treatment techniques for hazardous aniline-based organic contaminants

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Received: 8 July 2021 / Accepted: 10 May 2022 / Published online: 8 June 2022

Abstract

The growing contamination of various freshwater resources due to industrial effluent is a serious concern among the scientific community. Several organic compounds are essentially used as chemical intermediate in variety of industrial processes. These organic compounds are hazardous chemicals which are already considered dangerous to global public health and other forms of life due to their high toxicity, carcinogenicity. These organic contaminants are found present in the industrial effluents. Several treatment methods were applied in the literature for their elimination from wastewater to make their final disposal safe for environment. In this article, different kinds of physical, biological and advanced oxidation methods (AOPs) applied for the treatment of various important organic compounds were compared for their advantages and disadvantages. The results showed that the conventional treatment methods are not effective to treat these kinds of toxic and refractory chemical compounds. Therefore, AOPs were found to be the most promising treatment methods.

Keywords Advanced oxidation processes · Hazardous · Carcinogenicity · Organic compounds · Pharmaceuticals

Abbreviations

NIOSH	National Institute for Occupational Safety and Health
OSHA PEL	Occupational Safety and Health Administration Permissible Exposure Limit
IDLH	Immediately Dangerous to Life or Health
TWA	Time-weighted average
AOPs	Advanced oxidation processes
IARC	International Agency for Research on Cancer

Introduction

Water is a precious commodity for each living organism on the planet Earth. Though earth has large reservoirs of water in the form of oceans (70%), snow ice caps and glaciers (3%), the water actually available for human use is only 1% of total available water and is continuously getting lesser and lesser due to the contamination from various point (Zhou et al. 2016) and nonpoint sources (Ma et al. 2018) of water pollution. The point sources are the bulk contributors in polluting water resources day by day (Yang et al. 2016; Barilari et al. 2020). They include various industries and sewage treatment plants which directly discharge their effluent in environment. Due to the growing concern for the contamination of water bodies and their consequential effect on humans and other forms of life, these industrial discharges are regulated and checked by various federal and government agencies. Hence, industries must treat their effluents to meet the safe limits of several important water parameters and removal of harmful contaminants before disposal into freshwater resources. Therefore, the need of simple, effective and low-cost treatment techniques arises to minimize the burden of effluent treatment, to efficiently remove the harmful contaminants and to increase the overall productivity of industries (Rajasulochana and Preethy 2016). Various organic contaminants are such compounds of serious concern to the industries due to their potential detrimental effect on humans and other forms of life (Sang et al. 2019; Dewage et al. 2019; Chaturvedi and Katoch 2020a). In this study, I have tried to compare several treatment technologies to find out the most efficient and cost-effective treatment technology for organic contaminants.

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Aniline-based organic contaminants

There are several important but toxic organic compounds such as anisidine, aminobenzene, nitro aniline and pyridine which are still being used, produced and are present in the effluent of various industries. Most commonly and widely used organic compounds are either aniline itself or its derivatives. Aniline, also called as aminobenzene or phenylamine, is an aromatic organic compound with the chemical formula $C_6H_5NH_2$. An amino (–NH_2) group is attached to the benzene ring; hence, it is a prototypical aromatic amine (Bolt et al. 2016). It has an unpleasant rotten fish like odor like the other volatile amines (Podkościelny and László, 2007). Aniline readily catches fire and starts burning with smoky flames which is a characteristic of all the aromatic hydrocarbons. Aniline is basic in nature and appears as colorless oily liquid. Aniline is easily soluble in variety of solvents such as cold/hot water, diethyl ether and methanol (European Commission 2004). Aniline is required as an essential intermediate in the preparation of many organic substances such as fuel and rubber additives, corrosion inhibitors, azo dyes, antioxidants, pharmaceuticals, pesticides and antiseptics (Edalli et al. 2018; Bose et al. 2016; Wang et al. 2007). The primary use of aniline is in production of polyurethane precursors. The major use of aniline is to prepare methylene diamine and related organic compounds through condensation with formic aldehyde. The diamines thus produced undergo condensation with phosgene to give methylene diphenyl diisocyanate. The rest of the aniline is used to produce chemicals for rubber processing (9%), dyes and pigments (2%) and for herbicides (2%). Diphenylamine and phenylenediamines are aniline derivatives and act as antioxidants in when added to rubber. Aniline is also used in the preparation of paracetamol (acetaminophen), a common drug used to relieve fever. In dye industries, aniline is mainly used to prepare Indigo dye, which makes the jeans blue. Polyaniline, a polymer cable of conducting electricity intrinsically, is also a product of aniline.

Aniline is established as one of the highly toxic organic compounds (Hussain et al. 2014; Jagtap and Ramaswamy 2006). Aniline can be a potential mutagen and carcinogen; therefore, IARC has categorized it into group 2B (IARC 2012). Aniline has a water solubility of 3.4 g/100 mL, but the hydrochloride of aniline can dissolve up to 100 g/100 mL in water. Industrial wastewater containing aniline can be harmful to harms aquatic biomes due to its high toxicity and incalcitrant structure. NIOSH has considered aniline as a possible occupational carcinogen in humans and confirmed its carcinogenicity in animals (Pohanish 2017; Wang et al. 2016; Okazaki 2001). The NIOSH IDLH value and OSHA PEL value for aniline are 100 ppm and 5 ppm (19 mg/m^3), respectively (Pendergrass 1994). Aniline exposure can occur by dermal adsorption, inhalation and ingestion (Korinth et al. 2012, 2008, 2007). Its direct contact can result in skin burns and severe irritation to permanent damage of eyes. Acute exposure of aniline can decrease the potential of oxygen carrying and absorbing capacity of blood which may result in breathing difficulty, collapsing and finally death of exposed person (Pohanish 2017).

There are also vast variety of other organic compounds like aniline which are of prime concerns to various federal and local government authorities. Some of them are anisidines, nitroaniline, phenols, methylaniline, aminophenol, etc. which are some of the important chemicals used in several industries and are found in their wastewater. These contaminants are infamous for their carcinogenicity, toxicity and adverse effects on aquatic life and human being as well (Larrañaga et al. 2016; European Commission 2002; Budavari et al. 1996). The important regulatory parameters for different aniline-based organic compounds are shown in Table 1.

Table 1 clearly shows the threshold limit of various regulatory parameters, exposure after which can result in serious health effects for living organisms. The smaller the value of regulatory parameter for the organic compounds, the more dangerous it is for the living organisms. The OSHA PEL value and NIOSH REl value is smallest for o-Anisidine and p-Anisidine making them most dangerous among other organic compounds shown in the Table 1.

Therefore, several treatment methods have been applied for the removal of these kinds of organic compounds from the industrial effluents before their disposal into the environment. These contaminants have been treated and eliminated by various technologies including physical, biological and AOPs (Chaturvedi and Katoch 2020a; Bajpai et al. 2019). In this study various physical, biological and AOPs related methods used for treatment of wastewater containing these types of contaminants are reviewed and discussed for their merits and demerits.

Removal technologies for aniline-based organic contaminants

Aromatic aniline-based organic compounds have been treated from effluents by electrolysis (Li et al. 2017), photodecomposition (Pirsaheb et al. 2017), ozonation (Faria et al. 2007), biodegradation (Huang et al. 2018; Dino et al. 2019) and resin adsorption (Chen et al. 2020; Li et al. 2020). Complete decomposition has not been achieved by activated sludge processes, and their incalcitrant nature can prevent the biodegradation of several other harmful chemical species in wastewaters. Most of the physical methods like adsorption

Table 1 Hazard regulatory parameter for several aniline-based organic compounds

Organic compounds	IDLH value	OSHA PEL	NIOSH REL	IARC	Prime use
Aniline	100 ppm	19 mg/m^3 TWA (skin)	–	Group 3	Pharmaceuticals, dyes, pigments
o-Anisidine	50 mg/m^3	0.5 mg/m^3 TWA (skin)	0.5 mg/m^3 TWA (skin)	Group 2B	Pharmaceuticals and dye
p-Anisidine	50 mg/m^3	0.5 mg/m^3 TWA (skin)	0.5 mg/m^3 TWA (skin)	Group 3	Pharmaceuticals and dye
p-Nitroaniline	300 mg/m^3	6 mg/m^3 TWA (skin)	3 mg/m^3 TWA (skin)	Group 3	Dye industry
o-Toluidine	200 mg/m^3	22 mg/m^3 TWA (skin)	–	Group 1	Pesticides and dyes
Phenol	250 ppm	19 mg/m^3 TWA (skin)	19 mg/m^3 TWA (skin)	Group D	Resins and Nylon
Nitrobenzene	200 ppm	5 mg/m^3 TWA (skin)	5 mg/m^3 TWA (skin)	Group 2B	Dyes, drugs, Pesticides, etc
2-Aminopyridine	5 ppm	2 mg/m^3 TWA (skin)	2 mg/m^3 TWA (skin)	Group D	Pharmaceuticals
N, *N*-Dimethylaniline	100 ppm	25 mg/m^3 TWA (skin)	25 mg/m^3 TWA (skin)	Group 3	Dyes and pigments
p-Phenylene diamine	25 mg/m^3	0.1 mg/m^3 TWA (skin)	0.1 mg/m^3 TWA (skin)	Group 3	Permanent hair dye
Nitraamine	750 mg/m^3	1.5 mg/m^3 TWA (skin)	1.5 mg/m^3 TWA (skin)	–	Explosives
1,4-Diaminobenzene	25 mg/m^3	0.1 mg/m^3 TWA (skin)	0.1 mg/m^3 TWA (skin)	Group 3	Polymer, dyes and Kevlar
Cresols (o/m/p)	250 ppm	22 mg/m^3 TWA (skin)	10 mg/m^3 TWA (skin)	Group C	Chemical intermediate ad herbicides

Source NTP (2016), IARC (1999) and Barsan (2007)

are found to be very sensitive to pH of the wastewater. Other methods like incineration and ultrafiltration are not economical (Tanhaei et al. 2014; Shi et al. 2014), and thermal incineration can also cause air pollution (Sänger et al. 2001; Bie et al. 2007). Biological methods are eco-friendly techniques to destroy contaminants and transform them into non-toxic forms by natural means (Padoley et al. 2011; Jianping et al. 2006). However, for highly toxic and incalcitrant contaminant like aniline-based compounds, direct subjection to biological treatment can be troublesome. Therefore, chemical pretreatment by AOPS can be a suitable alternative as they increase the biodegradability and also minimize the toxicity of these compounds for microorganisms (Padoley et al. 2011; Chen et al. 2007). In recent researches, AOPs have surfaced as favorable technologies to destroy various harmful organic compounds in wastewater and reduce their toxicity and refractory nature. Various physical, biological and AOPs related methods used for treatment of aniline-based compounds are discussed below.

Physical treatment methods

Physical treatment methods such as membrane filtration, thermal incineration and adsorption have been used for the elimination of aniline-based organic compounds from wastewater. They have been shown to be treated with adsorption by resins (Gu et al. 2008; Jianguo et al. 2005), activated carbon (Valderrama et al., 2010), carbon nanotubes (Yan et al. 2011; Xie et al. 2007) and zeolites (O'Brien et al. 2008). Despite the low efficiency of adsorption system, the regeneration of adsorbent is also troublesome and incurs to additional cost (O'Brien et al. 2008, 2004). Aniline was also treated by thermal incineration in some studies but incineration is an energy intensive process with high fuel consumption and if incomplete combustion takes place, secondary pollution can happen due to the release of nitrogenous oxides (NO*x*) into the atmosphere as a by-product (Crini and Lichtfouse 2019; Sänger et al. 2001). Several membranes such as liquid emulsion membrane (Datta et al. 2003), silicone membrane (Sawai et al. 2005), reverse osmotic membranes (Gómez et al. 2009), nanofiltration (Shao et al. 2013) and ultrafiltration (Tanhaei et al. 2014) membranes were also applied in the treatment of aniline compounds. Although membrane filtration processes were found to be effective, complex rejection mechanism, incomplete removal and regular cleaning of the membrane by backwashing are several associated drawbacks (Hidalgo et al. 2011; Bellona et al. 2004; Drewes et al. 2003). Membrane fouling over time is also a significant limitation of this technology (Jhaveri and Murthy 2016; Cui et al. 2016). Different physical treatment technologies available for aniline-based organic compounds are summarized in Table 2.

Table 2 summarizes the physical treatment methods applied for the degradation of organic contaminants. Physical methods like adsorption, membrane filtration and thermal incineration were applied for these compounds. Although physical processes were found effective in the degradation of these compounds, they have certain major drawbacks like high energy cost, fouling of membrane and creation of more toxic secondary pollution.

Biological treatment methods

A variety of unicellular and multicellular organisms have been reported for the treatment of wastewater containing aniline-based organic compounds via biological methods (Arora 2015; Gharibzahedi et al. 2014; Liang et al. 2005). Aniline was shown to be treated by several fungi

Table 2 Physical treatment methods for organic contaminants

Physical treatment applied	Sub-category/material used	Chemicals concerned	Experimental condition	Major findings	References
Adsorption	Cu^{2+}-treated chitosan/alumina nanocomposite	aniline, oxalic acid and *N*,*N*-dimethyl aniline (NDA)	pH = 7.2, Adsorbent = 0.05 g volume = 50 mL Max, Ini. Conc. = 200 mg/L t = 4 h	Modified Chitosan was found more efficient than neat chitosan. 84 mg/g aniline and 61 mg/g NDA adsorption	Zavareh et al. (2017)
	Granular activated carbon	Aniline	Adsorbent = 10 g/L, Vol. = 100 mL, [Aniline] = 5.37 mmol/L and 4.54 mmol/L	0.73 mmol/g and 0.95 mmol/g adsorption of aniline on catechol and resorcinol systems, respectively	Suresh et al. (2012)
	Activated carbon (AC) and modified polymeric resin (MN200)	Mixture of aniline and phenol	Adsorbent = 0.5 g [Solute] = 50 mg dm^3 /0.5 m^3, t = 110 min	AC showed faster extraction of Aniline and Phenol than MN200	Valderrama et al. (2010)
	Cobalt-supported pumice	Anisidine	Adsorbent = 1 g, t = 48 h [Volume] = 20 cm^3	Cobalt supported pumice was found more efficient than pure pumice	Bardakçı et al. (2013)
	bifunctional polymeric resin	Aniline and 4-methylaniline (4MeA)	Adsorbent = 0.1 g [volume] = 100 mL, $[\text{Aniline/4MeA}]_i$ = 10.0 mmol/L	0.96 mmol and 1.24 mmol/mL of resin for Aniline and 4-Methylaniline, respectively	Jianguo et al. (2005)
	Activated carbon (AC) and modified bentonite (HDTMA)	Aniline	pH = 10, HDTMA = 30 g AC = 4 g [volume] = 100 mL, $[\text{Aniline}]_i$ = 300 mg/L	9.46 mg/g adsorption by modified Bentonite and 75.76 mg/g for AC	Tarlani et al. (2016)
	Activated Carbon (AC) coated with Chitosan	Aniline	Adsorbent = 0.2 g AC/chitosan = 0.5 [volume] = 50 mL, $[\text{Aniline}]_i$ = 20–50 mg/L, t = 100 min	Higher removal of aniline obtained for AC coated with Chitosan at wide pH range	Liu et al. (2015)
	Organo-zeolite (OZ)	Aniline and Nitrobenzene (NB)	pH = 7.5–8.5 Adsorbent = 300 g, $[\text{Aniline/NB}]_i$ = 30 mg/L, t = 15 h	Adsorption capacity of 2.36 mg/g and 3.25 mg/g of OZ obtained for Aniline and NB, respectively	Ersoy and Çelik (2004)
	Activated carbon	p-Nitroaniline (PNA)	pH = 7.6, $[\text{PNA}]_i$ = 200 mg/L	406 mg/g Adsorption capacity found	Salam et al. (2015)
	Nanocrystalline hydroxyapatite (HAP)	Nitrobenzene (NB)	pH = 2–11, Adsorbent = 5–20 g/L, $[\text{NB}]_i$ = 10 mg/L	Decrease in pH and temperature resulted in high adsorption of Nitrobenzene. 95% removal	Wei et al. (2010)

Table 2 (continued)

Physical treatment applied	Sub-category/material used	Chemicals concerned	Experimental condition	Major findings	References
Membrane filtration	Liquid emulsion membrane	Aniline	$[\text{Feed}]_i = 0.001$ M, Feed: Emulsion = 12:1 $t = 45$ min	98.53% removal of aniline	Datta et al. (2003)
	Micellar enhanced ultrafiltration	Aniline	pH = 7, $[\text{Aniline}]_i = 5$ ppm Volume = 300 mL Effective surface area = 40 cm^2	Approximately 80% removal of aniline	Tanhaei et al. (2014)
	Micellar enhanced ultrafiltration	Aniline	pH = 5 $[\text{Aniline}]_i = 1.5$ mM pressure = 3.5 bar	78.36% removal of aniline	Fu et al. (2017)
	silicone rubber membrane	Aniline and Phenol Derivative [APD]	$[\text{APD}]_i = 0.1$ mmol/L Volume = 200 mL Effective surface area = 9.1 × 10–4 m^2	Successful recovery of Aniline and Phenol from aqueous mixtures	Sawai et al. (2005)
	Nanofiltration	Aniline blue and Safranin O	pH = 11	More than 90% removal of both Aniline blue and Safranin O	Shao et al. (2013)

and microorganisms such as *Candida tropicalis* (Wang et al. 2011), *Candida albicans* (Jianping et al. 2006), *Pseudomonas* sp. (Jiang et al. 2016), *Delftia* sp. (Sheludchenko et al. 2005), *Acinetobacter* sp. (Takeo et al. 2013) and *Pigmentiphaga daeguensis* (Huang et al. 2018). Different biological treatment methods for aniline-based organic compounds are summarized in Table 3. Recently, several studies on microbial fuel cells for the treatment of these kinds of wastewater have also resulted in energy production (Singh and Dharmendra 2020; Zhang et al. 2019). Biological methods utilizes natural pathways in order to treat the wastewater for achieving the requisite wastewater quality, which makes them the most eco-friendly technique, but they become impractical when organic compounds of incalcitrant nature and high toxicity are to be treated by biological means (Padoley et al. 2008; Gotvajn and Zagorc-končan 2005).

Industrial wastewater contains a vast variety of harmful organic compounds and their treatment to achieve the mandatory effluent standards is problematic with biological methods. The main limitations of biological methods are: difficulty to grow and maintain culture in pure form; it requires longer time to stabilize the microorganisms and also for the oxidation of organic pollutants (Neyens and Baeyens 2003; Tony et al. 2012). It is very important to monitor and maintain healthy environmental condition for the proper growth of microorganism daily. Biological methods are the best eco-friendly practices and found efficient in the destruction of various organic compounds. Although the efficacy of biological methods is determined by the nature of available substrate to be acted upon by microbial enzymes (Karigar and Rao 2011), these processes are ineffective in the case of toxic contaminants with higher COD values than BOD (De Morais and Zamora 2005; Martınez et al. 2003). AOPs can be a good pretreatment options to address these limitations earlier to biological methods as they can improve the biodegradability by reducing the toxicity of organic contaminant non-specifically to a great extent (Padoley et al. 2011; Kavitha and Palanivelu 2004). Biological and physical treatment methods were found effective in the treatment of organic contaminants, but have certain disadvantages like high energy requirements, secondary pollution, cleaning and maintenance, slower elimination rate, etc. These problems can be overcome by AOPs as they have shown to be more successful and effective in the removal of similar organic compounds having high toxicity and incalcitrant nature.

Table 3 shows that a variety of biological processes have been applied for the effective elimination of these organic compounds. They seemed to be the eco friendliest as they utilize natural pathways for the degradation of organic compounds. But biological processes have many treatment constraints like selectiveness of microorganisms, organic compounds and also the need of large time duration for the effective removal of these kind of toxic compounds.

Table 3 Biological treatment methods for organic contaminants

Microrganism/ Method	Type of treatment	Chemicals concerned	Experimental conditions	Major findings	References
Delftia sp. XYJ6	Aerobic	Aniline	pH = 7, $T = 30$ °C [Aniline]$_i$ = 2000 mg/L, $t = 22$ h	*Delftia* sp. XYJ6 showed capability of Aniline conversion to intermediates	Liang et al. (2005)
Dietzia natronolimnaea[20]	Aerobic	Aniline	pH = 8, [Aniline]$_i$ = 300 mg/L, $t = 120$ h, $T = 30$ °C	87% removal of aniline	Gharibzahedi et al. (2014)
Pigmentiphaga daeguensis	Aerobic	Aniline	pH = 7, $t = 15$ h, [Aniline]$_i$ = 10 mg/L, $T = 30$ °C	100% removal of aniline	Huang et al. (2018)
Moving bed biofilm reactors	Aerobic	Aniline, para-Diaminobenzene [PDB] and para-Aminophenol [PAP]	[COD]$_i$ = 100–3500 mg/L, $t = 3$ days	COD removal of 90%, 87% and 75% for Aniline, [PDB] and [PAP]	Delnavaz et al. (2008)
Activated sludge process	Aerobic and Anerobic	Several Sulfonated Aromatic amines	pH = 7,	Only 2 and 4-Aminobenzene sulfonic acid showed removal of less than 1.8 g/L/day. And other tested compounds showed no sign of bio degradation	Tan et al. (2005)
Sequencing batch reactor system	Aerobic and Anerobic	Reactive Red (Azo dye)	pH = 7.9, [Dye]$_i$ = 20–50 mg/L, volume = 10 L	90% color removal, COD removal 88%	Koçyigit and Ugurlu (2015)
Batch column reactor	Aerobic and anerobic	Mordant Dye	pH = 8, [Dye]$_i$ = 4000 mg/L, volume = 1 L, $T = 23$ °C	61% dye mineralization, 70% aromatic amines removal obtained	Yan et al. (2018)

Therefore, as suggested and investigated by several researchers, pretreatment with AOPs in order to reduce the toxicity of these recalcitrant compounds can be a possible solution for their successful elimination from wastewater.

Advanced oxidation processes

AOPs have proved beneficiary in the treatment of several toxic and non-degradable compounds like aromatic organic compounds, pharmaceuticals, pesticides, petroleum wastewater, dyes and other refractory chemicals (Shahidi et al. 2015; Ribeiro et al. 2015; Gadipelly et al. 2014; Diya'uddeen et al. 2011). AOPs utilize the high oxidizing power of powerful oxidizing agents for the removal of organic compounds and were effectively used to remove recalcitrant organic contaminants from wastewater. They can destroy organic compounds by chemical and photochemical oxidation in the vicinity of a catalyst (Padoley et al. 2011; Kavitha and Palanivelu 2004). AOPs rely on the in situ generation of strong oxidants to eliminate organic compounds (Miklos et al. 2018; Bolton et al. 2001, 1996). Most of the AOPs utilize oxidizing species like HO·, but some can be based on other oxidizing species like sulfate and chlorine radicals (Miklos et al. 2018). HO· with 2.80 eV as oxidation potential surpasses most of the other oxidizing agents, and its reaction rate constants are much more higher than other methods like ozonation. HO· is unstable and highly reactive species in nature which must be generated continuously in situ by several means (Zhu et al. 2012; Esplugas et al. 2002; De Laat and Gallard 1999).

There are several AOPs available such as UV/TiO$_2$ catalysis, Fenton's oxidation, photo-Fenton oxidation, Fenton-like oxidation solar photo-Fenton, electro-Fenton oxidation and titanium dioxide-assisted photolysis (Jain et al. 2018; Boczkaj and Fernandes 2017; Asghar et al. 2015). Aniline-based organic compounds are shown to be treated and destroyed successfully by AOPs. Removal of acetanilide, p-nitroaniline and p-aminophenol using solar photo-Fenton and UV photo-Fenton oxidation was investigated by some researchers and revealed that both the treatment methods were more advantageous than the classic Fenton process due to their better oxidation ability, broader pH tolerance and lower Fe^{2+}

Table 4 Advanced oxidation processes for organic contaminants

Applied AOPS	Type of solution	Chemicals concerned	Experimental condition	Major findings	References
Hydrogen peroxide/UV light [H_2O_2/UV]	Synthetic Solution containing dyes	Monochlorotriazine associated Dyes	pH = 3, UV light, H_2O_2 = 680 mg/L	30.4% TOC removal	Alaton et al. (2002)
	Synthetic dye solution	Sulfur dye	pH = 7.29, UV = 54 W, $[Dye]_i$ = 70 mg/L H_2O_2 (3.9%) = 10 cm^3	Complete decolorization 93% TOC removal	Amin et al. (2008)
	Synthetic solution	Azo dye: CI reactive Red	pH = 8, UV = 60 W, H_2O_2 = 300 mg/L $[Dye]_i$ = 100 mg/L	97% dye removal and 35–40% COD removal in 30 min	Sudarjanto et al. (2005)
	Synthetic solution	2,4-Dinitroanisole (DNAN)	pH = 7, H_2O_2 = 1500 mg/L, [DNAN] = 250 mg/L	99.6% removal in 3 H 30% TOC removal	Su et al. (2019)
	Deionized water	Several Azo dyes	pH = 5.3, $[Dye]_i$ = 20 mg/L, H_2O_2 = 240 mg/L	95% color removal in less than 2 h	Shu et al. (2005)
	Deionized water	Mixture of Chlorophenoxy acids	pH = 7, UV light, H_2O_2 = 170 mg/L	56–79% COD removal in < 2 h	Fdil et al. (2003)
Ozone/UV light [O_3/UV]	Synthetic Solution	Deltamethrin, Triadimenol and Lambda-cyhalothrin	pH = 7, $[C]_i$ = 100 mg/L, UV light, flow rate = 1.2 g/h	92–96% chemical removal	Lafi and Al-Qodah (2006)
	Synthetic solution and real wastewater	p-nitroaniline (PNA)	pH = 9, $[PNA]_i$ = 10 mg/L, O_3 Supply = 0.9 g/h	94% PNA removal from synthetic solution and 81% removal in real wastewater	Rajabizadeh et al. (2019)
	Laboratory-scale washwater effluent	nitrosamines, amines and nitramines	pH = 8, UV = 272–537 mJ/cm^2, O_3 Supply = 21 mg/L	90% amine removal	Dai and Mitch (2015)
	Synthetic Solution	Aniline Aerofloat (AAF)	pH = 7, UV = 40 W O_3 Supply = 0.62–1.27 mg/min/L	99.84% AAF removal	Fu et al. (2019)
Titanium dioxide/UV light [TiO_2/UV]	Synthetic Solution	3-Chloropyridine (3-CLP)	pH = 6.8, [TiO_2] = 700 mg/L, $[3\text{-CLP}]_i$ = 40 mg/L, UV light	100% TOC removal in 300 min	Ortega-Liebana et al. (2012)
	Petroleum Refinery Wastewater	Aniline	pH = 12, [TiO_2] = 60 mg/L, $[Aniline]_i$ = 50 mg/L, T = 323 K, UV light	85% or more TCOD removal	Shahrezaei et al. (2012)
	Synthetic Solution	o-chloroaniline (OCA)	pH = 7–14, [TiO_2] = 100 mg/L, $[OCA]_i$ = 50 mg/L, UV light	Optimal TiO_2 concentration was 0.1 g/L	Choy and Chu (2005)

Table 4 (continued)

Applied AOPS	Type of solution	Chemicals concerned	Experimental condition	Major findings	References
Photo-Fenton $H_2O_2/Fe^{2+}/UV$	Synthetic textile wastewater	R94H reactive dye + PVA	pH = 3–5, UV light, $[H_2O_2] = 100$ mg/L, $[Fe^{2+}] = 20$ mg/L $[Dye]_i = 60$ mg/L	96% color removal in 30 min and 36% COD removal in 60 min	Kang et al. (2000)
	Synthetic Solution	Acid Yellow 36 and Methyl Orange	pH = 2, UV light, $[H_2O_2] = 0.008$ mM, $[Fe^{2+}] = 0.0003$ mM $[Dye]_i = 50$ mg/L	Complete color removal in 75 min, 100% TOC removal in 180 min	Macías-Sánchez et al. (2011)
	Synthetic solution	3-Chloropyridine (3-CLP)	pH = 2.8, UV light, $[H_2O_2] = 8.8$ mM, $[Fe^{2+}] = 0.88$ mM, $[3\text{-CLP}]_i = 40$ mg/L, $T = 25$ °C	100% TOC removal in 60 min	Ortega-Liebana et al. (2012)
	Synthetic solution in methanol as solvent	p-Anisidine(PA)	UV = 200 W, $[H_2O_2] = 0.0225$ mM, $[Fe^{2+}] = 0.003$ mM, $[PA]_i = 0.0122$ mM,	Intermediate p-Nitroanisole is formed	Kumar and Kabra (2008)
	Synthetic aqueous solution	Phenol	pH = 3, UV = 450 W, $T = 50$ °C, $[H_2O_2] = 100$ mM, $[Fe^{2+}] = 1$ mM, $[TOC]_i = 40$ mg/L	90% TOC removal in 3 h	Will et al. (2004)
Fenton's Oxidation H_2O_2/Fe^{2+}	Synthetic Solution	Several organic compounds (OC)	$[H_2O_2]/[OC] = 0–50$ $[Fe^{2+}]/[OC] = 0.01–1$ $[OC] = 300$ mg/L	$[Fe^{2+}]$ and $[H_2O_2]$ dosage is important for Fenton's oxidation	Chamarro et al. (2001)
	Synthetic solution	p-anisidine (PA)	$H_2O_2/Fe^{2+} = 70:1$ $[PA] = 0.5$ mM pH = 2.5	88.95% PA removal, 76.43% COD removal	Chaturvedi and Katoch (2020b)
	Synthetic Solution	o-Anisidine (OA)	$H_2O_2/Fe^{2+} = 60:1$ $[OA] = 0.5$ mM, pH = 3	85% OA removal, 74% COD removal	Chaturvedi and Katoch (2020c)
	Synthetic solution	o-, m-, p-cresols	pH = 3.0, $[H_2O_2] = 31.64$ mM, $[Fe^{2+}] = 0.90$ mM for o and p and 0.72 mM for m-isomer, $[Cres.]_i = 200$ mg/L	82% cresol removal, COD removal trend m > p > o isomer	Kavitha and Palanivelu (2005)
	Synthetic solution	Orange G (Azo dye)	$[H_2O_2] = 0.01$ mM, $[H_2O_2]/[Fe^{2+}] = 286:1$ $[Dye]_i = 1.11 \times 10^{-4}$ mM	94.6% decolorization within 60 min	Sun et al. (2009)
	Synthetic effluents	Phenolic Acids	pH = 3, $[H_2O_2] = 488$ mM, $[H_2O_2]/[Fe^{2+}] = 271$ mg $[TOC]_i = 370$ mg C/L	Final TOC = 123 mgC/L, final COD = 180 mgO_2/L final $BOD_5 = 146$ mgO_2/L	Martins et al. (2010)

Table 4 (continued)

Applied AOPS	Type of solution	Chemicals concerned	Experimental condition	Major findings	References
Fenton-like oxidation	Synthetic Solution	Methylene blue (MB)	pH = no constraint $[H_2O_2] = 0.5$ mL $[Fe_2SiS_4] = 0.1$ g, $[MB]_i = 5$ g/L Xenon Lamp: 300 W	85% degradation of MB and 96.9% degradation when illuminated	Shi et al. (2016)
	Synthetic Solution	2-Methoxyaniline (2MA) and 4-Methoxyaniline (4MA)	$H_2O_2/Fe^{3+} = 70:1$ [2MA/4MA] = 0.5 mM pH = 3 for 2MA pH = 2.5 for 4MA	83% 2MA removal and 86% 4MA removal with 71% and 72% COD removals, respectively	Chaturvedi and Katoch (2020d)
	Synthetic solution	p-nitroaniline (PNA)	$[H_2O_2]/[Fe^{3+}] = 100$ $[Fe^{3+}] = 0.05$ mM, pH = 3, $[PNA]_i = 0.5$ mM	laterite lacks by only 2–3% approx. in removal efficiency than $FeSO_4$	Amritha and Manu (2016)
	Synthetic solution	Malachite green dye (MGD)	pH = 3, $[H_2O_2] = 5 \times 10^{-2}$ M $[Fe^{2+}] = 1.0 \times 10^{-3}$ M $[MGD]_i = 3 \times 10^{-5}$ M	95% decolorization and 70% mineralization of MGD	Hashemian (2013)
	Synthetic Solution	Azo dye (Orange 7)	pH = 5.5, $[H_2O_2] = 9$ mL $[Fe_3O_4\text{-}MnO_2] = 600$ mg/L, $[Dye]_i = 50$ mg/L $T = 303$ K	96.8% removal of dye in 120 min	Fang et al. (2017)
Ultrasound/Ozone [US/O_3]	Synthetic Solution	Aniline	pH = 7, US density: 0.1 W/mL, O3 = 2 mg/min Aniline = 100 mg/L	82% removal of aniline and 20% TOC removal within 5 min	Song et al. (2007)
Photo-Fenton + Aerobic Batch reactor	Synthetic wastewater	Aniline	pH = 3.5, $[Fe^{2+}] = 15–40$ mg/L, $[H_2O_2]/[Fe^{2+}] = 20:1$ [Aniline] = 250 mg/L	94% or more removal of aniline and COD by combined process	Liu et al. (2012)
Ultrasound/Hydrogen peroxide US/H_2O_2	Synthetic solution	Aniline	pH = 3, $[H_2O_2] = 0.01$ mol/L [Aniline] = 20 mg/L US energy = 2.5 W/cm^2	95.91% removal of aniline in 45 min	Rahdar et al. (2019)
Photoassisted Electrochemical Incineration (PEI)	Synthetic solution	phenol, aniline, *n*-propanol and acetic acid	pH = 3.5, [Phenol/Aniline] = 5 M, UV light	UV light enhanced the oxidation	Treimer et al. (2001)
Heat-assisted persulfate oxidation	Synthetic Solution	Aniline	pH = 7, $[Aniline]_i = 0.50$ mL out of 0.09 g per100mL $T = 10$ to 50 °C	3 intermediates formed: nitrobenzene, 4-4′-diaminodiphenyl and 1-hydroxy-1,2-diphenylhydrazine	Xie et al. (2012)

requirement (Sheikh et al. 2008). In some researches, a combination of both photo-Fenton process and biological processes was studied for the degradation of aniline (Liu et al. 2012). This study showed that pH range of 3–4 and oxidation by photo-Fenton process increases the degradability of aniline by microorganisms. There are several studies available in the literature which demonstrated Fenton's reagent effectiveness in eliminating toxic organic contaminants from wastewater (Liu et al. 2012; Mingyu et al. 2011; Andreozzi et al. 1999). Electro-Fenton process is found more effective than fluidized bed Fenton process, but higher amount of H_2O_2 is required for electro-Fenton process, showing that fluidized bed Fenton process is more economical (Briones et al. 2012; Anotai et al. 2010). Aniline pretreatment by ozone followed by titanium dioxide photocatalysis showed overall increment in total organic carbon removal from the wastewater (Orge et al. 2017; Sanchez et al. 1998).

AOPs are influenced by several important reaction parameters such as solution pH, H_2O_2:Fe^{2+}, initial pollutant concentration. The pH of wastewater shown to increase the productivity of AOPs (Catalkaya and Kargi 2007). Normally acidic conditions are favored by AOPs resulting in faster degradation rather in alkaline conditions (Li et al 2015; Pera-Titus et al. 2004). The initial increase in Fe^{2+} results in higher amount of HO·, which further improves the degradation, until a critical concentration is reached after which the degradation is abruptly inhibited (Yilmaz et al. 2010). This can be explained by the fact that Fe^{2+} itself starts to absorb the HO· (Manu et al. 2011). Also, for higher initial pollutant concentration lower degradation is observed (Manu and Mahamood 2011). Various advanced oxidation processes applied for the treatment of aniline-based organic compounds are shown in Table 4.

Table 4 summarizes different kinds of AOPs applied for the treatment of organic compounds. There are several kinds of AOPs available in the literature. AOPs are faster and more effective than physical and biological processes. They have shown more than 90% removal in most of the degradation investigation. Sometimes 100% conversion of organic compound into carbon dioxide and water was also obtained. The simplest of all AOPs is Fenton oxidation which is the most eco-friendly of all other AOPs. Industries can opt for any of the AOPs as per their budget and requirement.

Discussion and conclusions

Aniline-based organic contaminants are found essential as chemical compounds for various industries. They have been identified by several governmental agencies as toxic, carcinogenic and mutagenic (Table 1). Therefore, their presence in the untreated effluents of these industries can be harmful and can cause serious adverse effects on humans as well as other forms of life. Through extensive studies of the literature, it was found that there are several treatment methods available for aniline-based organic compounds. There are variety of physical methods applied for the treatment of these compounds such as thermal incineration, membrane filtration and adsorption. The physical methods were found to be efficient and fast, but their limitations include formation of secondary air pollutants as in thermal incineration. In membrane filtration technologies, the consistent cleaning of the membrane by backwashing demands both energy and time, thereby adding costs. Moreover, fouling of membranes with time is a noteworthy drawback of this techniques. Biological processes are considered as the most eco-friendly technologies, but their effectiveness rely on the nature of the available substrate to be acted upon by microorganisms. Therefore, for toxic and incalcitrant organic contaminants, biological processes are not practicable. Moreover, biological process has other limitations like slower removal rate and requires continuous monitoring and maintenance. Due to these limitations, researches have moved toward AOPs, as they have advantages like non-specific pollutant degradation, faster removal rate, ease of operation and found eco-friendly and economical. AOPs also have certain limitations like pH dependence, sludge formation and complex reaction chemistry.

Acknowledgements The author is sincerely thankful to all the editors and reviewers for their suggestions for further refining the quality of this study.

Funding This research did not receive any specific grant from funding agencies in the public, commercial or not-for-profit sectors.

Availability of data and materials Not applicable.

Code availability Not applicable.

Declarations

Conflict of interest The authors declare that they have no competing interests.

Consent to participate Not applicable.

Consent for publication Not applicable.

Ethics approval Not applicable.

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