



# The utility of ultraviolet beam in advanced oxidation-reduction processes: a review on the mechanism of processes and possible production free radicals

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## Abstract

Advanced oxidation processes (AOPs) and advanced reduction processes (ARPs) are a set of chemical treatment procedures designed to eliminate organic (sometimes inorganic) contamination in water and wastewater by producing free reactive radicals (FRR). UV irradiation is one of the factors that are effectively used in oxidation-reduction processes. Not only does the UV beam cause the photolysis of contamination, but it also leads to the product of FRR by affecting oxidants—reductant, and the pollutant decomposition occurs by FRR. UV rays produce active radical species indirectly in an advanced redox process by affecting an oxidant ( $O_3$ ,  $H_2O_2$ ), persulfate (PS), or reducer (dithionite, sulfite, sulfide, iodide, ferrous). Produced FRR with high redox potential (including oxidized or reduced radicals) causes detoxification and degradation of target contaminants by attacking them. In this review, it was found that ultraviolet radiation is one of the important and practical parameters in redox processes, which can be used to control a wide range of impurities.

**Keywords** Ultraviolet beam · Advanced oxidation · Advanced reduction · Free radical

## Introduction

As it has become an important issue for human health, the management and treatment of wastewater have significantly increased in the modern era by inventing and using conventional wastewater treatment (CWWT). CWWTs are

classified as physical, chemical, and biological processes. The sequential or simultaneous use of these processes in combination may be an efficient way to degrade a resistant pollutant to CWWT that can be referred to as xenobiotics (anthropogenic chemicals that are toxic and dangerous). Therefore, tertiary or polishing methods are used to remove such pollutants, but limitations related to implementation in full scale and cost-benefit are still the notable factors limiting their practical use (Hossein Panahi et al. 2020).

Among the tertiary methods, AOPs and ARPs are technologies that have received much attention during the last several decades. AOPs are a set of chemical purification procedures designed to degrade organic (sometimes inorganic) materials in water and wastewater by producing free reactive radicals (FRR) and pollutants oxidizing through them. Therefore, AOPs are based on the in situ generations of strong oxidants, i.e., hydroxyl radicals ( $OH^\bullet$ ), sulfate ( $SO_4^{\bullet-}$ ), and superoxide ( $O_2^{\bullet-}$ ) radicals for the oxidation of organic pollutants (Khodadadi et al. 2020; Pandis et al. 2022; Saravanan et al. 2022). The simplest advanced oxidation process is the Fenton reaction, which is carried out by hydrogen peroxide along with an iron catalyst to produce  $OH^\bullet$  (Al-Musawi et al. 2019). Some of the AOPs use UV

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## Advanced oxidation processes based on UV

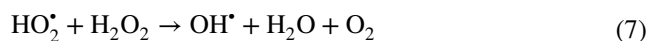
### UV-based oxidant

#### H<sub>2</sub>O<sub>2</sub> activation based on UV

Many AOPs use free radicals called highly reactive hydroxyl radicals (OH•) to reduce a wide range of organic pollutants in aqueous solutions. According to the literature, one of the most widespread UV/AOP processes is UV/H<sub>2</sub>O<sub>2</sub>, which is extensively studied due to its efficiency in producing FRR such as OH• (He et al. 2012). The UV/H<sub>2</sub>O<sub>2</sub> process is an attractive option for generating non-selective and highly reactive radicals. In the UV/H<sub>2</sub>O<sub>2</sub> process, H<sub>2</sub>O<sub>2</sub> is made photolysis by UV irradiation to produce OH• whose redox potential is 2.7 V and 1.8 V in acidic and neutral solutions. Since OH• is short-lived, it is produced in situ during ozone- and UV-based processes by activating stable precursors such as H<sub>2</sub>O<sub>2</sub> (Lee et al. 2020). The results of studies show that during the UV/H<sub>2</sub>O<sub>2</sub> process, photolysis of these oxidants occurs at a wavelength of  $\lambda = 254$  nm, which leads to the production of FRR such as OH• (Ike et al. 2018). Hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) is a beneficial oxidizer because it is an environmentally friendly oxidizer that only contains hydrogen and oxygen during decomposition (Zhao et al. 2021). The main disadvantage of hydrogen peroxide is its low oxidizing ability in environmental conditions, which must be activated to have proper performance. Moreover, to make photolysis, UV photons play a very important role in activating H<sub>2</sub>O<sub>2</sub> and producing FRR. Also, the H<sub>2</sub>O<sub>2</sub> dose directly affects the production of radicals, and thus the removal efficiency of pollutants will improve when the dose of hydrogen peroxide is optimal. The corresponding reactions in activating H<sub>2</sub>O<sub>2</sub> and producing FRR are stated below (Eqs. 4–8).

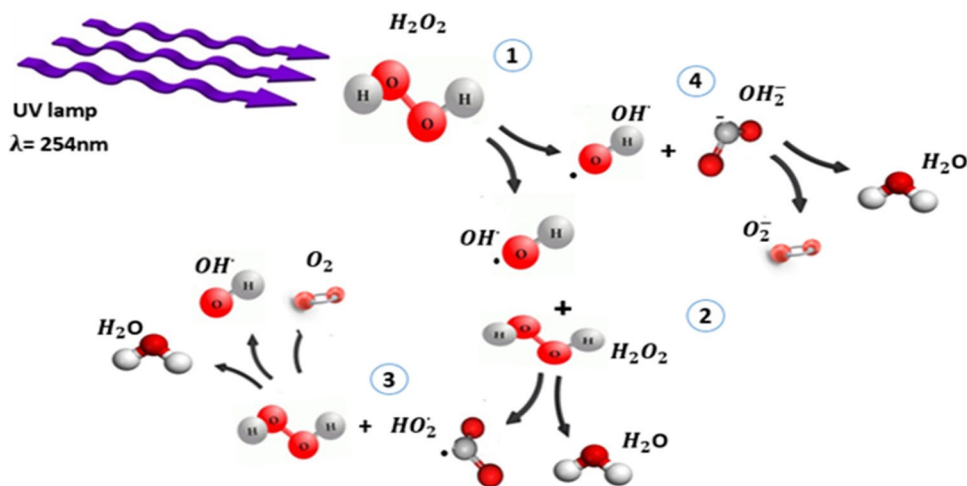
According to Eq. 4, H<sub>2</sub>O<sub>2</sub> changes into 2OH• by receiving UV photons (Eq. 4), and other H<sub>2</sub>O<sub>2</sub> molecules that have

not received UV photons convert into H<sup>+</sup> and HO<sub>2</sub><sup>-</sup> ionic forms (Eq. 5). Other related chain reactions are described in Eqs. 6–8.



Finally, OH• and HO<sub>2</sub><sup>•</sup> radicals are formed, which can play a role in the decomposition of organic substances. In addition to degrade organic pollutants, OH• can inactivate microorganisms, while HO<sub>2</sub><sup>•</sup> cannot have a noticeable effect on inactivating microorganisms (Zhao et al. 2018). The reaction of HO<sub>2</sub><sup>•</sup> with its reaction intermediates creates a scenario what H<sub>2</sub>O<sub>2</sub> excessive concentration can lead to the consumption of OH• (scavenger phenomena), and OH• reacts with radical scavenger faster than the target pollutants. The graphical schematic of the UV/H<sub>2</sub>O<sub>2</sub> process in producing FRR is shown in the second Fig. 2. UV/H<sub>2</sub>O<sub>2</sub> processes are used to eliminate a lot of contamination including Pb–EDTA, Rhodamine B, melatonin, antipyrine, benzophenone-3, ofloxacin, and acetamiprid, and some of them along with possible radicals produced are presented in Table 1. UV/H<sub>2</sub>O<sub>2</sub> is a fast process and has already been done on a large scale. Moreover, UV irradiation can serve as an effective disinfectant for a variety of microorganisms in addition to activate the H<sub>2</sub>O<sub>2</sub>. However, the residual hydrogen peroxide can be dangerous and can also have a negative impact on the process if not reduced. So, it may require removal downstream of the process (Chang et al. 2010; Hollman et al. 2020; Lee

**Fig. 2** Graphical schematic of radical production in UV/H<sub>2</sub>O<sub>2</sub>



**Table 1** Eliminated pollutants by UV/H<sub>2</sub>O<sub>2</sub> process along with free radicals

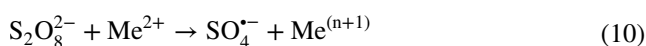
Contaminations	Process type	Radical species	Reference
Pb-EDTA	UV/H <sub>2</sub> O <sub>2</sub>	OH <sup>•</sup> , HO <sub>2</sub> <sup>•</sup> , O <sub>2</sub> <sup>•-</sup>	Jiraroj et al. (2006)
Rhodamine B	UV/H <sub>2</sub> O <sub>2</sub>	SO <sub>4</sub> <sup>•-</sup> , OH <sup>•</sup> , HO <sub>2</sub> <sup>•</sup> , O <sub>2</sub> <sup>•-</sup> , Cl <sup>•</sup> , Cl <sup>•-</sup> , HOCl <sup>•-</sup>	AlHamedi et al. (2009)
Melatonin	UV/H <sub>2</sub> O <sub>2</sub>	OH <sup>•</sup> , HO <sub>2</sub> <sup>•</sup>	Xu et al. (2009)
Antipyrine	UV/H <sub>2</sub> O <sub>2</sub>	OH <sup>•</sup> , HO <sub>2</sub> <sup>•</sup>	Tan et al. (2013)
Benzophenone-3	UV/H <sub>2</sub> O <sub>2</sub>	OH <sup>•</sup> , O <sub>2</sub> <sup>•-</sup> , O <sup>•-</sup>	Lee et al. (2021)
Ofloxacin	UV/H <sub>2</sub> O <sub>2</sub>	HO <sub>2</sub> <sup>•</sup> , OH <sup>•</sup> , HOCl <sup>•-</sup> , Cl <sup>•</sup> , Cl <sub>2</sub> <sup>•-</sup>	Liu et al. (2020)
Acetamidiprid	UV/H <sub>2</sub> O <sub>2</sub>	HO <sub>2</sub> <sup>•</sup> , OH <sup>•</sup> , O <sup>•-</sup>	Chen et al. (2018)

et al. 2020; Lee et al. 2021; Srithep and Phattarapattama-wong 2017; Ye et al. 2022).

### Persulfate activation based on UV

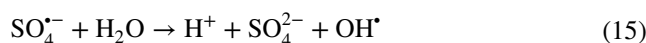
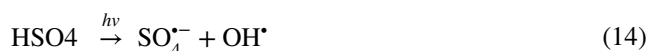
Persulfate-based advanced oxidation processes can be considered promisingly for the removal of stable micropollutants in water and wastewater treatment processes. The types of various persulfate (PS) species are peroxymonosulfate (PMS) and peroxy disulfate which can consider as stable oxidizers in solid state (compared to H<sub>2</sub>O<sub>2</sub>), cost-benefit (compared to ozone), high aqueous solubility (compared to permanganate), and high stability at the ambient environment (Wang et al. 2020).

Because PDS has a higher quantum efficiency (i.e., 0.7 mole/E for PDS and 0.5 mole/E for H<sub>2</sub>O<sub>2</sub>) and molar absorption coefficient than hydrogen peroxide (i.e., 21.1 M<sup>-1</sup> cm<sup>-1</sup> for PDS and 18.6 M<sup>-1</sup> cm<sup>-1</sup> for H<sub>2</sub>O<sub>2</sub>), so it has a higher photolysis rate (Ike et al. 2018). All the features of this material facilitate its transportation, storage, and use (Ding et al. 2020). Various persulfate species can be activated to produce FRR (SO<sub>4</sub><sup>•-</sup>) by several agents such as UV irradiation, heat, ultrasound radiation, and transition metal oxides (Eqs. 1–4) (Bu et al. 2017; Gao et al. 2022; Varank et al. 2020; Walde-mer et al. 2007).



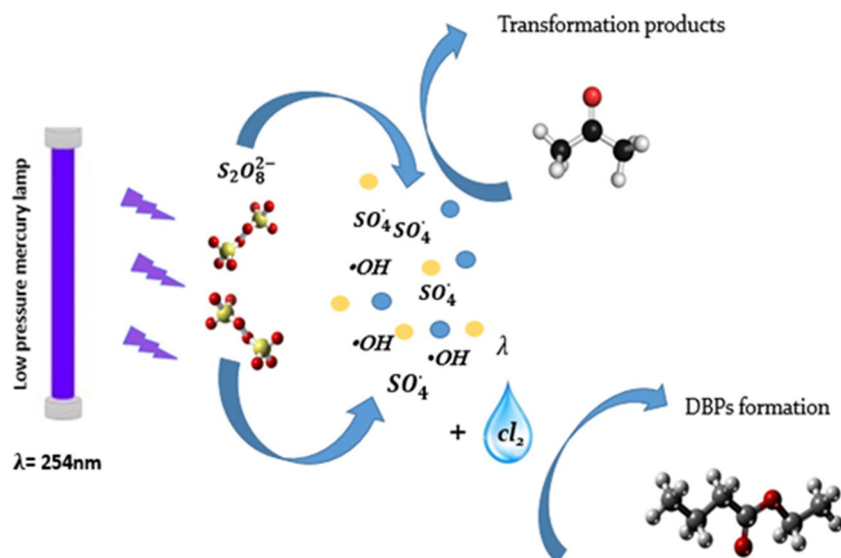
Many activators have been evaluated for persulfate activating and considering the effective factors in choosing an activator such as economic, energy consumption, and potential pollution of heavy metals. UV irradiation is the most suitable agent for environmental applications (Cui et al. 2022; Wang et al. 2020). It is considered an

environmentally friendly and efficient agent to activate an oxidant including PS and PMS which can generate SO<sub>4</sub><sup>•-</sup>. Na<sub>2</sub>S<sub>2</sub>O<sub>8</sub> and K<sub>2</sub>S<sub>2</sub>O<sub>8</sub> (UV/PS) are the most common persulfate compounds that are used to supply sulfate ions in the advanced oxidation process. The graphical schematic of the UV/PS process is shown in the second Fig. 3. SO<sub>4</sub><sup>•-</sup> by a high oxidation potential ( $E = 2.5\text{--}3.1$  V) is the strongest oxidant after OH<sup>•</sup> that can decompose organic substances, and it is more stable than OH<sup>•</sup> and acts selectively in a wide range of pH. Contrary to OH<sup>•</sup>, SO<sub>4</sub><sup>•-</sup> has a long half-life such it makes more efficient than OH<sup>•</sup>. SO<sub>4</sub><sup>•-</sup> and OH<sup>•</sup> are produced according to the following equations (Eqs. 13–15) (Cui et al. 2022; Fu et al. 2019; Wang et al. 2020):



Moreover, SO<sub>4</sub><sup>•-</sup> radicals have a longer lifespan (30–40 μs) compared to OH<sup>•</sup> (100 ns), which makes it more reactive to organic matter through the electron transfer mechanism (Hoang et al. 2022). They react with electron-donating groups such as hydroxyl (OH), alkoxy (-RO), and amino (-NH<sub>2</sub>) groups (Ou et al. 2017; Yu et al. 2021). SO<sub>4</sub><sup>•-</sup> tends to react with organic matter through an electron transfer mechanism rather than H<sup>+</sup> abstraction and is less affected by water matrix components such as natural organic matter (NOM) and alkalinity, indicating that SO<sub>4</sub><sup>•-</sup> oxidation may complete OH-based AOPs with diverse combinatorial reactivates, product patterns, and energy yields (Qin et al. 2020a). SO<sub>4</sub><sup>•-</sup> radicals prefer to attack nucleophile structures via electron transfer, which can degrade rapidly a wide range of organic contaminants (Liu et al. 2020) and is used to destroy various pollutants such as saccharin (Ye et al. 2022), ofloxacin (Liu et al. 2020), imidacloprid (Wang et al. 2020), and benzophenone-3 (Lee et al. 2021).

**Fig. 3** Graphical schematic of radical production in UV/persulfate



In a study conducted by Wang et al., the degradation pathway of imidacloprid is evaluated by  $SO_4^{\bullet-}$ , and degradation pathways were proposed based on the produced intermediates (Wang et al. 2020). They report that the UV/PS process under similar conditions results in better degradation than the UV/PMS process. To explain this phenomenon, by activating PS or PMS under UV radiation, the rate of radical production can be calculated. A parameter called extinction coefficient ( $\epsilon$ ) of PS or PMS in Eq. 18 can greatly affect the radical rate in similar conditions. According to the relevant calculations, the extinction coefficient calculated in the same conditions for PS was higher than PMS, so it can be said that the radical production rate in the UV/PS system ( $r_{\text{radicals PS}}$ ) was higher than in the UV/PMS system ( $r_{\text{radicals PMS}}$ ). The rate of radical production is calculated as follows:

$$\text{Radicals} = \phi I_0 f (1 - 10^{-A}) / V \quad (16)$$

$$A = b \left( \epsilon c + \sum_i \epsilon_i c_i \right) \quad (17)$$

$$f = \epsilon c b / A \quad (18)$$

Some previous studies on the removal of many pollutants including antipyrine, sulfadimethoxine, benzophenone-3, ofloxacin, halostonitrile, chloramphenicol, acetamiprid, and Cu (II)-EDTA by the method of UV/persulfate and some of them with possible radicals produced are presented in Table 2. UV/persulfate does not produce sludge similar to the heat/persulfate process and has been used on a large scale. However, the disadvantage of UV/persulfate is its inappropriate efficiency in the presence of turbidity, and the sulfate radical is quickly destroyed in alkaline environments. So UV/persulfate process is used in the tertiary stages of water purification.

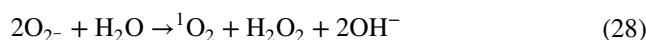
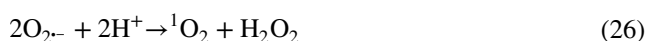
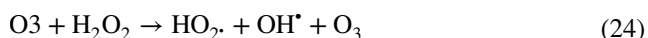
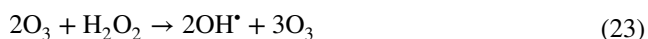
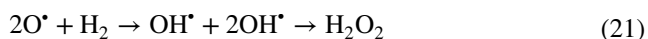
#### Ozone activation based on UV

Previous reports show that the addition of UV radiation to ozonation ( $O_3$ ) improves the pollutant degradation process, which is attributed to the synergistic effect between  $O_3$  and UV. Ozonation is one of the powerful treatment methods used

**Table 2** Eliminated pollutants by UV/PS process along with free radicals

Contaminations	Process type	Radical species	Reference
Antipyrine	UV/PS	$SO_4^{\bullet-}$ , $HO_2^{\bullet}$ , $OH^{\bullet}$	Tan et al. (2013)
Sulfadimethoxine	UV/PS	$SO_4^{\bullet-}$ , $OH^{\bullet}$ , $HCO_3^{\bullet}$ , $CO_3^{\bullet-}$	Shad et al. (2020)
Benzophenone-3	UV/PS	$SO_4^{\bullet-}$ , $OH^{\bullet}$	Lee et al. (2021)
Ofloxacin	UV/PS	$HOCl^{\bullet-}$ , $Cl^{\bullet}$ , $Cl_2^{\bullet-}$ , $OH^{\bullet}$ , $SO_4^{\bullet-}$ , $S_2O_8^{\bullet-}$	Liu et al. (2020)
Haloacetonitriles	UV/PS	$HOCl^{\bullet-}$ , $OH^{\bullet}$ , $Cl^{\bullet}$ , $Cl_2^{\bullet-}$	Hou et al. (2017)
Chloramphenicol	UV/PS	$OH^{\bullet}$ , $SO_4^{\bullet-}$ , $S_2O_8^{\bullet-}$ , $O^{\bullet-}$ , $NO_2^{\bullet}$ , $NO_3^{\bullet}$	Hou et al. (2017)
Acetamiprid	UV/PS	$OH^{\bullet}$ , $SO_4^{\bullet-}$	Chen et al. (2018)
Cu(II)-EDTA	UV/PS	$SO_4^{\bullet-}$ , $HO_2^{\bullet}$ , $OH^{\bullet}$ , $HCO_3^{\bullet}$	Xu et al. (2017)

to remove some pollutants from aqueous solutions. However,  $O_3$  alone cannot achieve complete mineralization of these pollutants (Chai et al. 2018).  $O_3$  is an oxidant that can play a role in the decomposition of pollutants both directly and indirectly. In direct mode,  $O_3$  with a relatively high redox potential affects various bands of organic compounds and detoxifies them. But in the indirect mode,  $O_3$  turns into an active radical species in the UV/ $O_3$  system and produces different types of reactive oxygen species such as superoxide radical ( $O_2^{\bullet-}$ ),  $OH^{\bullet}$ , and singlet oxygen ( $^1O_2$ ), and subsequently fast decomposition is its result (Wang et al. 2021a). The graphic representation of FRR in the UV/ $O_3$  process is shown in Fig. 4. In particular, the UV lamp provides the energy necessary to break the  $O_3$  chemical bonds within to produce reactive oxygen species with high reactivity (Eqs. 19–29) (Liu et al. 2021; Yang et al. 2020). In the synergistic reaction between  $O_3$  and UV beam, ozone is produced directly by Eqs. 19–21 and indirectly by Eqs. 22–29 (Emam 2012; Norabadi et al. 2020).



As we can find from the equations, the radicals including  $O^{\bullet}$ ,  $OH^{\bullet}$ ,  $HO_2^{\bullet}$ ,  $O_2^{\bullet-}$ , and  $HO_2^{\bullet-}$  product play a role in the decomposition of organic matter. The degradation pathway of ciprofloxacin (CIP) by possible radicals under the UV/ $O_3$  process has been investigated in a study conducted by Liu et al. (see Fig 5. supplementary). The two main active sites of ciprofloxacin were in the aromatic tertiary amine group and the secondary aliphatic amine group in the piperazine ring. The primary attack site of  $^1O_2$  was the piperazine ring, and with the attack of  $^1O_2$ , the hydroxyl present in the piperazine ring is removed, leading to the formation of CIPeI-3, CIPeI-5, CIP VI-1, and CIP-II-1. Cleavage of the aromatic ring of CIP-II-4 and CIP-II-5 led to the formation of CIP-1, which is attributed to electron transfer. In general, due to the attacking piperazine ring,  $^1O_2$  and  $O_2^{\bullet-}$  are processed to produce a keto derivative of the piperazine ring, pathway I, pathway II, pathway VI, and pathway VIII. Then,  $OH^{\bullet}$  attacks the C-C bonds, and CIP-III-1 is obtained. The anthranilic acid analog intermediate, CIP-III-2, is formed in the decarbonylation of CIP-III-1. Then, the quinolone group was cleaved by OH to produce CIP-III-3. In route IV,  $^1O_2$  attacks the cyclopropyl group ( $-C_3H_5$ ) and oxidizes to the carboxylic acid product (CIP-IV-1) (Liu et al. 2021). Table 3 shows a list of pollutants decomposed in the UV/ $O_3$  method including ciprofloxacin, organophosphorus pesticides, carbamate pesticides, carbamate pesticides, N-Nitrosopyrrolidine, 17-estradiol, bisphenol A, benzalkonium sulfuronoside, and possible radicals produced in this process.  $O_3$ /UV is more stoichiometrically efficient in generating  $OH^{\bullet}$  than  $H_2O_2$ /UV or  $H_2O_2$ / $O_3$ ; however, it is less energetically efficient than  $H_2O_2$ /UV or  $H_2O_2$ / $O_3$  for generating large quantities of  $OH^{\bullet}$  due to the low solubility of  $O_3$  in water compared to  $H_2O_2$ .

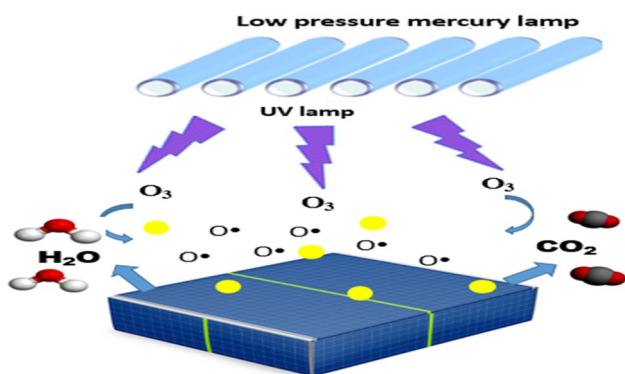


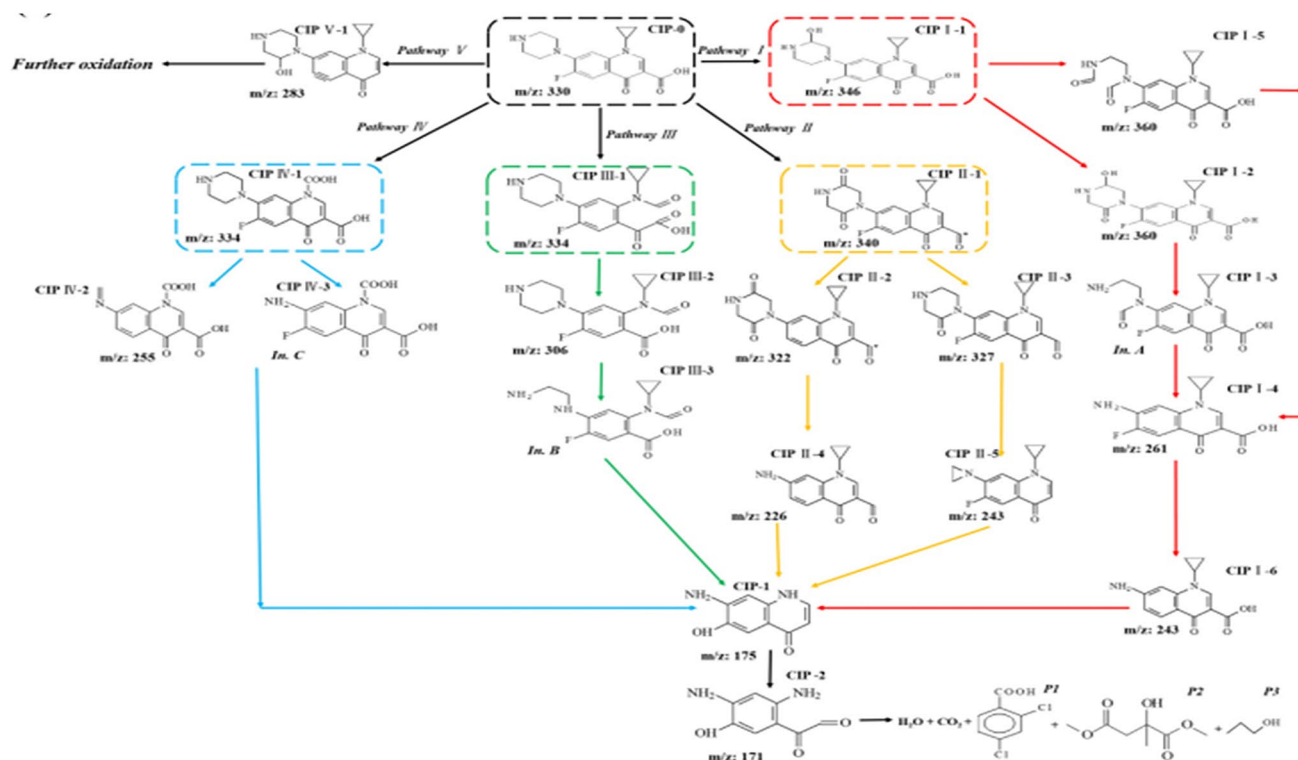
Fig. 4 Graphical schematic of radical production in UV/ $O_3$  process

## UV-based combination

Combined processes based on UV can be very diverse, and the reason for that is the key role of UV in AOPs, so that it cannot only be used alone but also play a multiple role in other complex combined AOPs. Below is an overview of the most likely AOPs UV-based.

### UV/ $H_2O_2$ / $O_3$ process

During previous reports, AOPs, especially  $O_3$  and their combination with  $H_2O_2$  to increase the formation of  $OH^{\bullet}$ , have



**Fig 5** Supplementary. Degradation pathway of ciprofloxacin (CIP) by possible radicals under UV/O<sub>3</sub> process

**Table 3** Eliminated pollutants by UV/O<sub>3</sub> process along with free radicals

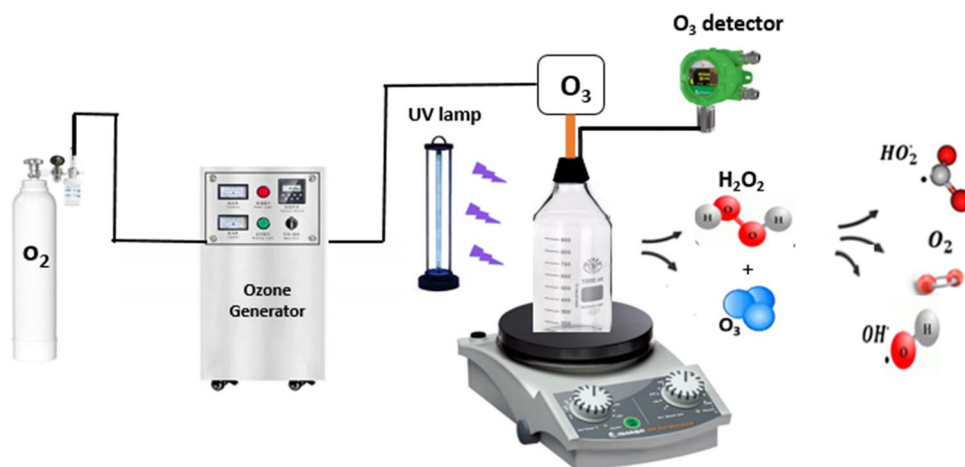
Contaminations	Process type	Radical species	Reference
Ciprofloxacin	UV/O <sub>3</sub>	HO <sub>2</sub> <sup>•</sup> , OH <sup>•</sup> , O <sub>2</sub> <sup>•-</sup> , HO <sub>2</sub> <sup>•</sup> , Cl <sup>•</sup> , Cl <sub>2</sub> <sup>•-</sup>	Liu et al. (2021)
Pesticides	UV/O <sub>3</sub>	OH <sup>•</sup> , HO <sub>2</sub> <sup>•</sup> , HO <sub>3</sub> <sup>•</sup> , O <sub>3</sub> <sup>•-</sup>	Samadi et al. (2010)
N-Nitrosopyrrolidine	UV/O <sub>3</sub>	OH <sup>•</sup> , O <sub>2</sub> <sup>•-</sup>	Chen et al. (2016)
17-Estradiol and bisphenol A	UV/O <sub>3</sub>	OH <sup>•</sup> , O <sub>2</sub> <sup>•-</sup> , O <sub>3</sub> <sup>•-</sup> , HO <sub>2</sub> <sup>•</sup> , HO <sub>3</sub> <sup>•</sup> , HO <sub>4</sub> <sup>•</sup>	Irmak et al. (2005)
Benzalkonium chloride	UV/O <sub>3</sub>	OH <sup>•</sup> , HO <sub>2</sub> <sup>•</sup> , O <sub>2</sub> <sup>•-</sup>	Wang et al. (2021a)
Quinoline	UV/O <sub>3</sub>	OH <sup>•</sup> , HO <sub>2</sub> <sup>•</sup> , O <sub>2</sub> <sup>•-</sup> , O <sub>3</sub> <sup>•-</sup>	Wang et al. (2004)
Linuron	UV/O <sub>3</sub>	OH <sup>•</sup> , O <sub>2</sub> <sup>•-</sup> , O <sub>3</sub> <sup>•-</sup> , HO <sub>2</sub> <sup>•</sup>	Rao and Chu (2009)
Sulfosalicylic acid	UV/O <sub>3</sub>	OH <sup>•</sup> , HO <sub>2</sub> <sup>•</sup> , O <sub>3</sub> <sup>•-</sup>	Tong et al. (2005)

been shown as a sustainable and efficient approach (Boczkaj and Fernandes 2017). The graphical abstract of this system is depicted in Fig. 6. O<sub>3</sub> can provide efficient degradation, but it is not affordable and cost-effective as well, and cost can be a limitation in these methods (Tollefsen et al. 2014). Also, hydrogen peroxide is a very popular oxidizing agent because it is a green and environmentally friendly chemical, such that water and oxygen are the only end products. In the UV/O<sub>3</sub>/H<sub>2</sub>O<sub>2</sub> system, the amount of ozone consumption is reduced by adding UV; as a result, the degradation cost is reduced, and the oxidation capacity of the system is improved for decomposition of organic compounds. In other words, the simultaneous presence of both O<sub>3</sub> and H<sub>2</sub>O<sub>2</sub> oxidants during UV irradiation has a synergistic effect (Arslan et al. 2017).

Due to the coexistence of several oxidizing species with different reactivity and selectivity (H<sub>2</sub>O<sub>2</sub>, O<sub>3</sub>, OH<sup>•</sup>, HO<sub>2</sub><sup>•</sup>), a rapid removal of a pollutant can occur (Bavasso et al. 2020). In the presence of UV rays, H<sub>2</sub>O<sub>2</sub> and O<sub>3</sub> decompose to O<sub>2</sub> and HO<sub>2</sub><sup>•</sup> radicals (Eq. 27). OH<sup>•</sup>, which reacts quickly with organic substances, also is produced from the reaction between O<sub>3</sub> and HO<sub>2</sub><sup>•</sup> (Eq. 29) (Oturán and Aaron 2014). The chain reactions that explain the breakdown of O<sub>3</sub> and H<sub>2</sub>O<sub>2</sub> are summarized below (Eqs. 27–34):



**Fig. 6** Graphical abstract of radical production in UV/H<sub>2</sub>O<sub>2</sub>/O<sub>3</sub> system

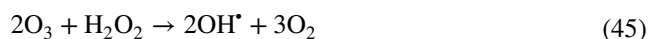
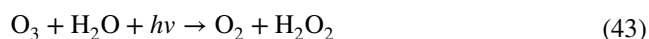
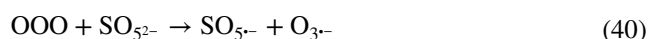
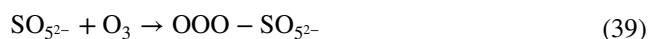


As can be seen from the reactions (Eqs. 27–34), OH• and HO<sub>2</sub>• radicals are formed, and they play a role in the decomposition of pollutants depending on their redox potentials. A list of pollutants degraded in the UV/H<sub>2</sub>O<sub>2</sub>/O<sub>3</sub> process including ortho-toluidine, furfural, azo dye AR88, winery wastewater, and possible radicals produced in Table 4 is stated. UV/H<sub>2</sub>O<sub>2</sub>/O<sub>3</sub> process is more effective than UV/H<sub>2</sub>O<sub>2</sub> and UV/O<sub>3</sub> processes in eliminating organic substances due to the production of more free radicals. However, more materials and energy are consumed in the UV/H<sub>2</sub>O<sub>2</sub>/O<sub>3</sub> process. In addition, the variety of free radicals is greater in the UV/H<sub>2</sub>O<sub>2</sub>/O<sub>3</sub> process, which is considered one of its capabilities (Arslan et al. 2017; Boczkaj and Fernandes 2017).

**UV/O<sub>3</sub>/persulfate system**

In recent years, the structural diversity of micro-pollutants in different water matrices makes measuring their reduction efficiency has led to challenges for the general design of AOPs. In the UV/O<sub>3</sub>/PS process, UV activates the molecule O<sub>3</sub> to OH• and PMS to SO<sub>4</sub>•<sup>-</sup>, which can be said that the

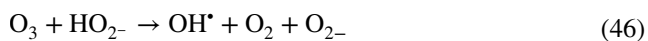
produced radicals (OH• and SO<sub>4</sub>•<sup>-</sup>) are relatively stronger than the original molecules (O<sub>3</sub> and PMS) (Yu et al. 2021). The various complex reactions in which FRR is produced are mentioned below (Eqs. 35–46):



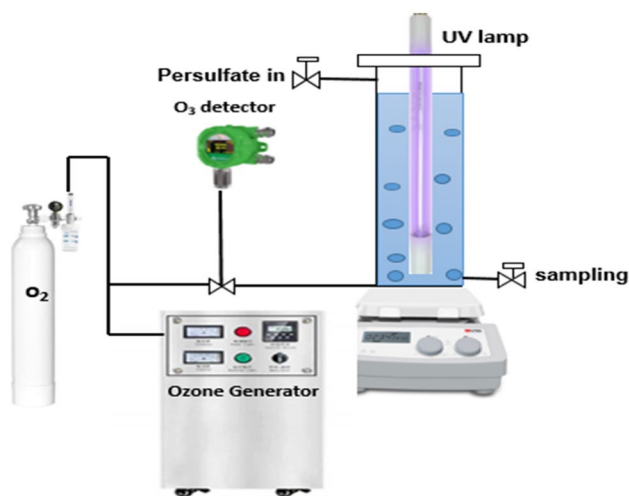
**Table 4** Eliminated pollutants by UV/H<sub>2</sub>O<sub>2</sub>/O<sub>3</sub> process along with free radicals

Contaminations	Process type	Radical species	Reference
Ortho-toluidine	UV/H <sub>2</sub> O <sub>2</sub> /O <sub>3</sub>	OH•, HO <sub>2</sub> •, HO <sub>3</sub> •, O <sub>2</sub> • <sup>-</sup> , O <sub>3</sub> • <sup>-</sup>	Shokri et al. (2016)
Furfural	UV/H <sub>2</sub> O <sub>2</sub> /O <sub>3</sub>	OH•, HO <sub>2</sub> •, Cl•, O <sub>2</sub> • <sup>-</sup> ,	Tang et al. (2011)
Azo dye AR88	UV/H <sub>2</sub> O <sub>2</sub> /O <sub>3</sub>	OH•, HO <sub>2</sub> •, O•	Yang et al. (2013)
Winery wastewater	UV/H <sub>2</sub> O <sub>2</sub> /O <sub>3</sub>	OH•, HO <sub>2</sub> •, HO <sub>3</sub> •, O <sub>2</sub> • <sup>-</sup> , O <sub>3</sub> • <sup>-</sup> , O•	Lucas et al. (2010)





As can be seen in chain reactions, the possible free radicals produced in the UV/O<sub>3</sub>/PS process include OH<sup>•</sup>, SO<sub>5</sub><sup>•-</sup>, SO<sub>4</sub><sup>•-</sup>, and O<sub>3</sub><sup>•-</sup>. The FRR will play a role in the decomposition of organic pollutants due to their redox potential. Figure 7 shows the general schematic of free radical production in the UV/O<sub>3</sub>/PS system. UV/O<sub>3</sub>/persulfate process to remove various pollutants such as atrazine, Reactive Red 120 dye, clofibric acid, atenolol, diclofenac, and possible radicals produced in these processes is presented in Table 5. The most important feature of UV/O<sub>3</sub>/persulfate is the simultaneous production of two valuable radicals, SO<sub>4</sub><sup>•-</sup> and OH<sup>•</sup>, which cause the process to be highly efficient in eliminating pollutants in both acidic and alkaline environments. Moreover, considering the presence of ozone and persulfate oxidants, it can be noted that the most diverse free radicals are in the UV/O<sub>3</sub>/persulfate process and UV/H<sub>2</sub>O<sub>2</sub>/persulfate (Yu et al. 2021). But UV/O<sub>3</sub>/persulfate and UV/H<sub>2</sub>O<sub>2</sub>/persulfate processes, like most advanced oxidation processes, are used for the tertiary stages of water and wastewater treatment because they are not affordable for the primary and secondary stages of purification.



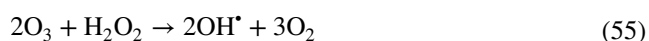
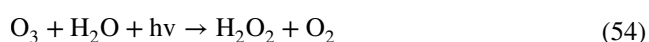
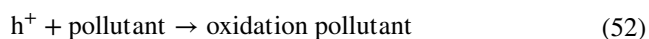
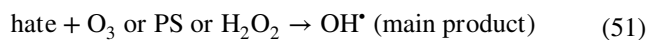
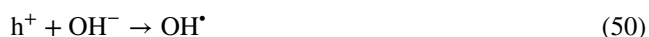
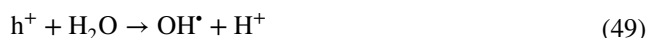
**Fig. 7** Graphical abstract of radical production in UV/O<sub>3</sub>/PS system

## UV/oxidizing/photocatalyst system

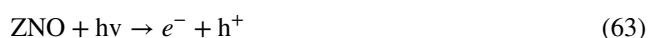
Oxidation processes are based on the production of FRR, destroying a wide range of resistant and toxic organic pollutant (aromatic and aliphatic) compounds quickly and without selectivity and the production of free radicals in the processes that are auxiliary oxidizers. Like UV, it depends on ozone or special catalysts like titanium or zinc oxide (Wang and Wang 2018). The use of ultraviolet rays with a suitable oxidizing agent such as hydrogen peroxide or ozone is very effective for removing all types of resistant organic pollutants to biological decomposition. One of the most important combined processes based on ultraviolet rays is the UV/oxidizing/photo catalyst system. The basis of the UV/oxidizing/photocatalyst process is the production of FRR, which enables it to react quickly with organic compounds in aqueous solution. In this process, ultraviolet rays have multiple roles, and they can decompose the pollutant through three mechanisms. The first mechanism is the direct decomposition of the pollutant by ultraviolet rays, but this mechanism is not very likely due to the limitation in matching the wavelength of the pollutant with different bands in the pollutant, and it can be said that about 5% of the pollutant is decomposed by this mechanism. However, the highest percentage of pollutant removal in UV/H<sub>2</sub>O<sub>2</sub>/photocatalyst or UV/O<sub>3</sub>/photocatalyst systems is related to the next two mechanisms. In the second mechanism, UV affects the oxidizing substance (O<sub>3</sub>, H<sub>2</sub>O<sub>2</sub>, PS) and produces various radical species, causing the degradation of organic substances. In addition, the third mechanism is the effect of ultraviolet rays on the semiconductor material in the reactor, which is known as the photocatalytic process. The simultaneous presence of oxidizing and activating agents together in the UV/oxidizing/photocatalyst process leads to abundant and diverse production of free radicals with high redox potential, and their synergistic effect leads to the removal of organic substances with high efficiency (Aboudalle et al. 2018; Vo et al. 2019; Wang and Wang 2018). During these processes, the pollutants in water are completely decomposed into carbon dioxide and water molecules or other harmless substances under the influence of the synergistic effect in the reactor (Jahantigh et al. 2019; Malakootian et al. 2019; Mashayekh-Salehi and Moussavi 2016; Mehrjouei et al. 2015). The responsible chain reactions in the system are mentioned below (Eqs. 47–56):

**Table 5** Eliminated pollutants by UV/O<sub>3</sub>/PS process along with free radicals

Contaminations	Process type	Radical species	Reference
Atrazine	UV/O <sub>3</sub> /PS	SO <sub>4</sub> <sup>•-</sup> , OH <sup>•</sup>	Qin et al. (2020b)
Reactive Red 120 dye	UV/O <sub>3</sub> /PS	SO <sub>4</sub> <sup>•-</sup> , OH <sup>•</sup>	Sharma et al. (2015)
Clofibric acid	UV/O <sub>3</sub> /proxymonosulfate	SO <sub>4</sub> <sup>•-</sup> , OH <sup>•</sup> , ClOH <sup>•-</sup> , Cl <sup>•-</sup> , Cl <sub>2</sub> <sup>•-</sup>	Qin et al. (2020a)
Atenolol	UV/O <sub>3</sub> /proxymonosulfate	SO <sub>5</sub> <sup>•-</sup> , SO <sub>4</sub> <sup>•-</sup> , OH <sup>•</sup> , O <sub>3</sub> <sup>•-</sup> , CO <sub>3</sub> <sup>•-</sup> , Cl <sup>•</sup> , HCO <sub>3</sub> <sup>•-</sup> , O <sup>•-</sup>	Yu et al. (2021)
Diclofenac	UV/O <sub>3</sub> /PS	SO <sub>4</sub> <sup>•-</sup> , OH <sup>•</sup> , HO <sub>2</sub> <sup>•</sup> , O <sub>2</sub> <sup>•-</sup>	Jabbari et al. (2020)



In addition to ultraviolet rays, oxidizing agents ( $\text{O}_3$ ,  $\text{H}_2\text{O}_2$ , PS) can also decompose pollutants through different routes. There are two effective ways in the degradation of contaminants by oxidizing agents. First, a pollutant is directly oxidized to less toxic molecules by electrophilic attacks of the oxidizing agent; in this route, ozone has only a relative efficiency (Eqs. 47–56). Another way is the decomposition of the oxidizing agent to FRR and its attacks on the pollutant during a series of indirect oxidation reactions (Eqs. 57–65) (Dong et al. 2022; Mehrjouei et al. 2015).



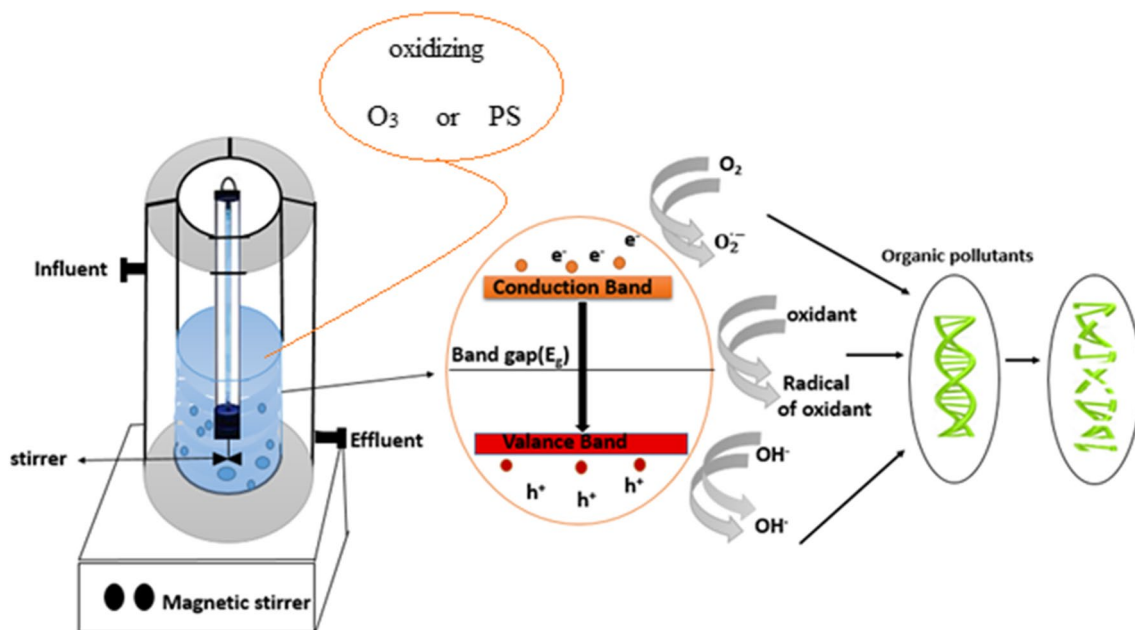
In the process of photocatalytic ozonation, the synergistic effects of absorption, photolysis, photo-catalysis ozone, and the production of hydroxyl radicals occur mostly in a short reaction time. The effectiveness of ozone is increased when combined with processes such as UV light, catalysts, and hydrogen peroxide. Plus, a catalyst like UV plays the role of activation for the oxidant if the catalyst used in this system is not a semiconductor material, and the general corresponding reaction is stated below:



According to Eq. 66, FRR (such as  $\text{SO}_4^{\bullet-}$ ) forms during chain reactions. The general schematic of FRR production for the UV/ $\text{O}_3$ /PS system is shown in Fig. 8. In the process of UV/oxidizing/photocatalyst system, various pollutants including amoxicillin, ampicillin, and cloxacillin antibiotics, real textile effluents, textile dyestuff, polyvinylpyrrolidone have been removed, which these compounds and also the radicals formed during these processes are shown in Table 6. The important point in the UV/oxidizing /photocatalyst process is that in addition to do photocatalytic reactions, the catalyst and UV simultaneously play a role in activating the oxidant, and it is considered one of the prominent features of the process. Here, there is a high diversity of active and radical species as well (Kurniawan et al. 2006; Zhang and Chu 2022).

## UV-based photocatalyst

The photocatalytic process is one of the AOPs that has been widely investigated to remove various compounds resistant to biological degradation on a laboratory and pilot scale in the last decade. It is known as environmentally friendly, sustainable, and economical technology. In this process, a semiconductor material ( $\text{ZnO}$ ;  $\text{TiO}_2$ ;  $\text{WO}_3$ ;  $\text{SnO}_2$ ; etc.) under UV radiation is used in both homogeneous and heterogeneous forms. In the UV/photocatalyst process, UV is not considered an activating agent, but its interaction with the catalyst leads to the production of free radicals. A graphical schematic of FRR production in a photocatalytic process is depicted in Fig. 9 (Guo et al. 2017; Khalid et al. 2017; Koe et al. 2020). The main characteristic of a semiconductor is the bandwidth of conductivity. The area between these two bands is known as the band gap. By irradiating UV to the catalyst, the absorption of photons having higher energies than that of the band gap leads to the transfer of an electron from the valence band to the conduction band, with the simultaneous production of gaps in the valence band (reaction 67). These gaps have a very high oxidation potential which is sufficient to produce hydroxyl radicals from water molecules and hydroxide ions adsorbed onto the semiconducting surface (reactions 68 and 69). The formed electrons can react with

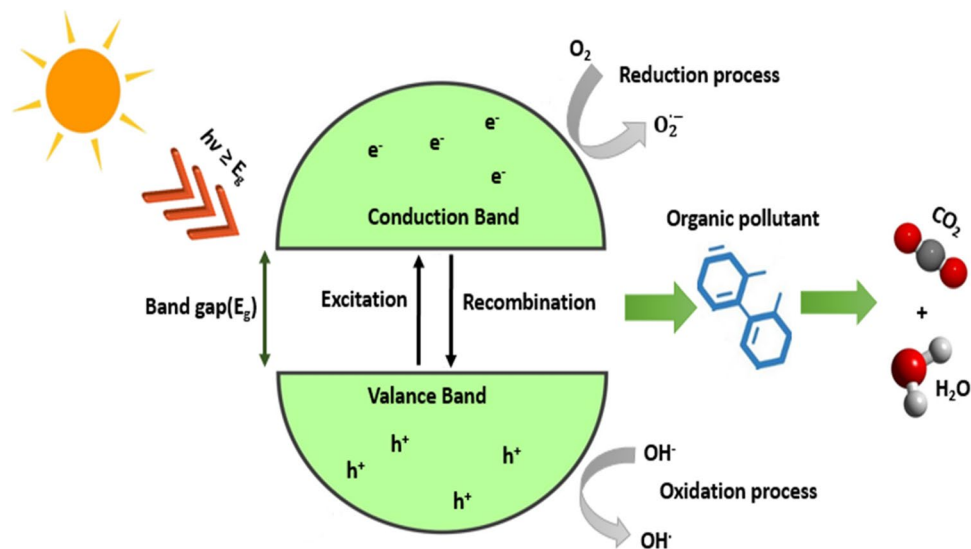


**Fig. 8** Graphical abstract of radical production in UV/oxidizing/photocatalyst system

**Table 6** Eliminated pollutants by UV/oxidizing/photocatalyst system along with free radicals

Contamination	Process type	Radical species	Reference
Amoxicillin, ampicillin, and cloxacillin antibiotics	TiO <sub>2</sub> /H <sub>2</sub> O <sub>2</sub> /UV	OH•, O <sub>2</sub> • <sup>-</sup>	Elmolla and Chaudhuri (2010)
Real textile effluents	TiO <sub>2</sub> /H <sub>2</sub> O <sub>2</sub> /UV	OH•	Garcia et al. (2007)
Textile dyestuff	Fe(III)/H <sub>2</sub> O <sub>2</sub> /solar UV	OH•, R•, RO <sub>2</sub> •	Parilti and Akten (2010)
Polyvinylpyrrolidone	TiO <sub>2</sub> /H <sub>2</sub> O <sub>2</sub> /UV	OH•, R• <sup>+</sup> , O <sub>2</sub> • <sup>-</sup>	Suave et al. (2018)

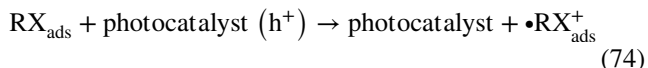
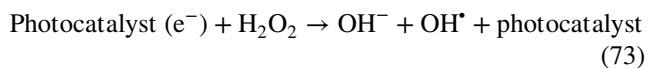
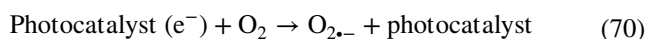
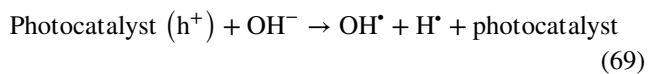
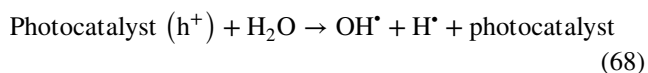
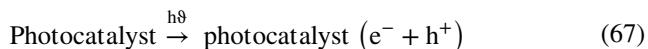
**Fig. 9** Graphical schematic of FRR production in photocatalytic reaction



the adsorbed oxygen molecule and turn it into a superoxide radical (O<sub>2</sub>•<sup>-</sup>) which in turn reacts with proton to form peroxide radicals (reactions 70–72). Further, hydrogen peroxide acts as

an electron receiver, and according to reaction 73, produces additional hydroxyl radicals. The adsorbed substrate (RX<sub>ads</sub>) may be directly oxidized by electron transfer (reaction 74)

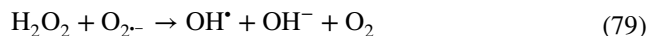
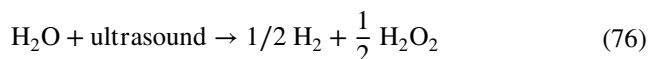
(Arghavan et al. 2021, Kamani et al. 2023, Khodadadi et al. 2020, Khodadadia et al., Nasseh et al. 2020a, Nasseh et al. 2020b).



As shown in the reactions, positive holes  $h^+$  are formed on the surface of the catalyst. They are very reactive, and when they come in contact with the pollutant, they decompose them. But the formed electron enters a chain of reactions, and  $\text{O}_2^{\bullet-}$ ,  $\text{HO}_2^\bullet$ ,  $\text{OH}^\bullet$ , and  $\bullet\text{RX}_{\text{ads}}^+$  radicals are formed. The most obvious difference between UV/photocatalyst process and other AOPs is producing  $h^+$ , which can convert non-biodegradable organic substances into low-risk or safe substances. However, its main failure is the reusability of a photocatalyst and its application in homogeneous aqueous beds (Guo et al. 2017, Khalid et al. 2017, Koe et al. 2020).

The photocatalytic process can be combined with other methods and factors such as ultrasound and heat. Ultrasound waves (US) in the water media during the cavitation phenomenon and the creation of cavities with high temperature and pressure can directly contribute to the production of  $\text{OH}^\bullet$  (Saalbach et al. 2018). However, the use of this type of radiation alone requires a lot of time and energy to destroy resistant organic compounds. Therefore, the US is used in combination with other methods, which can be referred to as UV/hate, US/photocatalytic, UV/catalyst processes,

etc. In the sonophotocatalytic process (US/photocatalytic), the following reactions may occur: (1) oncolysis of water, (2) reaction of  $\text{H}_2\text{O}_2$  with hydrogen atoms consisting, (3) photodecomposition of hydrogen peroxide, (4) reaction of hydrogen peroxide with the superoxide radical anion formed during photocatalysis, and (5) the reaction of  $\text{H}_2\text{O}_2$  with the conductive layer electrons (Eqs. 75–79). Several OH radicals usually recombine into  $\text{H}_2\text{O}$  and  $\text{H}_2\text{O}_2$  which are known as the scavenger phenomenon (Eqs. 80–83) (Norabadi et al. 2022; Saalbach et al. 2018).



$\text{H}^\bullet$ ,  $\text{HO}_2^\bullet$ , and  $\text{OH}^\bullet$  radicals are the most common radicals produced during the sonochemical process, which can also interact with other oxidants to produce  $\text{OH}^\bullet$  (see Eqs. 75–77). The UV-based photocatalyst process has been used to remove pollutants such as phenol, amoxicillin, ampicillin, cloxacillin, reactive brilliant blue KN-R, and organic pollutants, a list of which as well as the radicals produced in the removal process are shown in Table 7.

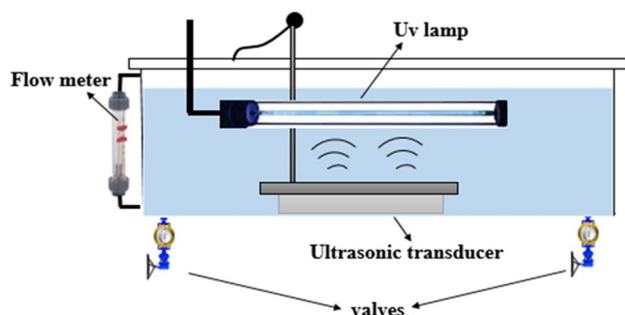
### UV/other activation agent

UV can also be used along with other activators (US, hate, catalyst) related to oxidizing compounds ( $\text{O}_3$ ,  $\text{H}_2\text{O}_2$ , PS). UV waves are one of the most important activators that are used

**Table 7** Eliminated pollutants by UV-based photocatalyst along with free radicals

Contaminations	Process type	Radical species	References
Phenol	UV/TiO <sub>2</sub>	OH <sup>•</sup> , OH <sup>•-</sup> , O <sub>2</sub> <sup>•-</sup> , HO <sub>2</sub> <sup>•</sup> , R <sup>•</sup>	Chiou et al. (2008)
Amoxicillin, cloxacillin	UV/TiO <sub>2</sub>	O <sub>2</sub> <sup>•-</sup> , OH <sup>•</sup>	Elmolla and Chaudhuri (2010)
Reactive brilliant blue KN-R	UV/TiO <sub>2</sub>	O <sub>2</sub> <sup>•-</sup> , OH <sup>•</sup> , HO <sub>2</sub> <sup>•</sup>	Liu et al. (2010)
Organic pollutants	UV/TiO <sub>2</sub>	O <sub>2</sub> <sup>•-</sup> , OH <sup>•</sup>	Poulopoulos et al. (2019)

together with UV. The US plays a role in the production of free radicals based on the cavitation phenomenon. Cavitation events are classified into two groups: stable and transient. However, in both production groups, a very local temperature of about 4000 K and a pressure of more than 1000 atmospheres are observed. The energy of the event depends on the cavitation intensity, which in turn can depend on many factors, including the amplitude and frequency of the sound wave (Saalbach et al. 2018). During the formation and collapse of bubbles, critical temperature and pressure conditions (about 5000 K and 500 atmospheres) are created, following this water sonolysis process, and then the formation of  $\text{H}^\bullet$  and  $\text{OH}^\bullet$  free radicals can happen as fast as oxidation. In contrast to other combined methods, the integration of UV with other activators aims to improve the process efficiency with the help of synergistic effects so that maximum efficiency can be achieved in the shortest time. A graphical schematic of FRR production in UV/other activation agents is observed in Fig. 10. In this route, UV only plays the role of direct pollutant decomposition, and the photolysis process occurs, which was mentioned earlier. The removal of pollutants including melatonin, sulfosalicylic acid, real textile effluents, and organic pollutants by methods including UV/Fe(II)/ $\text{H}_2\text{O}_2$ , UV/ $\text{O}_3/\text{TiO}_2$ , UV/Fe(II)/ $\text{H}_2\text{O}_2$ , and UV/ $\text{TiO}_2/\text{H}_2\text{O}_2/\text{Fe(III)}$  as well as radicals produced in these processes is shown in Table 8. These processes have been investigated more on a laboratory scale, and examining the efficiency of two electromagnetic activating agents (UV/US) is not very common due to high energy consumption. On the other hand, the use of UV and metal catalysts pointed out problems such as producing sludge and reducing the efficiency of ultraviolet rays in activating the oxidant by absorbing the metal catalyst.



**Fig. 10** Graphical schematic of FRR production in UV/US

**Table 8** Eliminated pollutants by UV/other activation agent along with free radicals

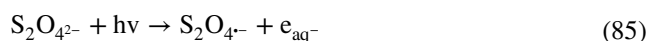
Contaminations	Process type	Radical species	Reference
Melatonin	UV/Fe(II)/ $\text{H}_2\text{O}_2$	$\text{HO}_2^\bullet$ , $\text{OH}^\bullet$	Xu et al. (2009)
Sulfosalicylic acid	UV/ $\text{O}_3/\text{TiO}_2$	$\text{HO}_3^\bullet$ , $\text{OH}^\bullet$	Tong et al. (2005)
Real textile effluents	UV/Fe(II)/ $\text{H}_2\text{O}_2$	$\text{OH}^\bullet$	Garcia et al. (2007)
Organic pollutants	UV/ $\text{TiO}_2/\text{H}_2\text{O}_2/\text{Fe(III)}$	$\text{HO}_2^\bullet$ , $\text{OH}^\bullet$	Pouloupoulos et al. (2019)

## Advanced reduction processes (ARPs)

Recently, a new group of water and wastewater treatment methods called advanced reduction processes (ARPs) has been proposed and is being developed on a laboratory scale. AOPs, ARPs, and combined advanced oxidation/reduction processes (AO/RP) are highly efficient technologies in the elimination of emerging micro-pollutants from water and wastewater. Almost all types of aqueous solution pollutants, including pesticides, PPCPs (pharmaceuticals and personal care products), dyes, plasticizers, and toxic metals are successfully removed from aqueous solution systems using various AO/RPs. All the AO/RPs rely on the generation of free radicals/species (i.e.,  $\bullet\text{OH}$ ,  $\text{SO}_4^{\bullet-}$ ,  $e_{\text{aq}}^-$ ,  $\text{SO}_3^{\bullet-}$ ,  $\text{H}\bullet$ ), which react with the target contaminants, resulting in their decomposition. A redox reaction includes AOP and ARP. Therefore, there is an ARP along with the AOP, and it is used to remove reduced contamination. An ARP decomposes oxidized pollutants by generating highly reactive reducing radicals (RRR) through a combination of reagents and activation agents (see general reactions in Fig. 11). Important activators that have been mentioned in various studies are UV beams and US waves. In this paper, an overview of UV processes has been studied; therefore, ARPs are classified into several categories below.

### UV/dithionite

Dithionite ions ( $\text{S}_2\text{O}_4^{2-}$ ), a powerful reducing agent, are encouraging reductants for application in ARPs, as they have a weak S-S bond which is readily cleaved into sulfur dioxide radical anions ( $\text{SO}_2^{\bullet-}$ ) by activation methods. The dithionite absorbs UV radiation at a wavelength of 315 nm when UV collides with dithionite ions, and the result is the production of reduced  $\text{SO}_2^{\bullet-}$  radicals (see Eqs. 84, 85). In fact, ultraviolet rays by affecting the S-S bond and breaking it into two parts produce radicals including sulfur dioxide ion, dithionite ion, and hydrate electron.



Both dithionite and sulfur dioxide radicals are strong reductants. The literature has reported that the standard reduction potential of sulfur dioxide radical and dithionite

is  $-0.66\text{ V}$  and  $-1.12\text{ V}$  (at pH 7 and  $25\text{ }^\circ\text{C}$ ) respectively. UV/dithionite system can be utilized for the elimination of dyes, refractory organic compounds in the petrochemical industry, antibiotics, and nitrates. The graphical schematic of RRR production in the UV/dithionite process is displayed in Fig. 12.

Some of it convert to other products (bisulfite, metabisulfite, thiosulfate, and trithionate) when dithionite enters an aqueous solution, and they are responsible for pollutant decomposition. The next factor in the decomposing of organic pollutants is reduced radicals caused by the impact of UV on dithionite. However, pollutant decomposition in the UV/dithionite process will occur in an acidic aqueous bed (Jung et al. 2018a; Jung et al. 2018b).

It has been reported in several studies that the UV/dithionite process has been used to eliminate organic contaminants such as arsenite, acid yellow 17, and selenite, and the summary

of possible radicals produced is reported in Table 9. In these studies, the photolysis of dithionite and the efficiency of the produced radicals in pollutant removal have been evaluated.

### UV/sulfite

Both UV and sulfite ( $\text{SO}_3^{2-}$ ) are commonly used in wastewater treatment disinfection; UV is a standalone process, and sulfite is a dechlorination chemical. Nevertheless, when these two factors are used together, an advanced reduction process takes place. Sulfite can strongly absorb UV beams at  $254\text{ nm}$  and pH 10 with a molar absorption coefficient of  $18.2\text{ M}^{-1}\text{ cm}^{-1}$ . Initially, the result of the interaction between UV and sulfite is the production of  $e_{\text{aq}}^-$  and  $\text{SO}_3^{\bullet-}$  which is presented in Eq. 86. In UV/sulfite, ultraviolet rays excite the electron located in the S-O bond to create the sulfide radical, and other radicals are created in a chain of reactions.

Fig. 11 Advanced reduction processes in an overview

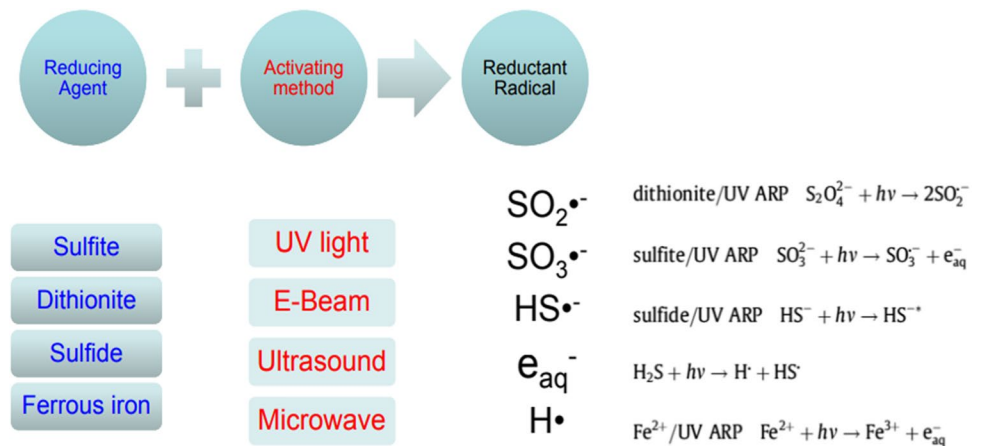


Fig. 12 Graphical schematic of RRR production in the UV/dithionite process

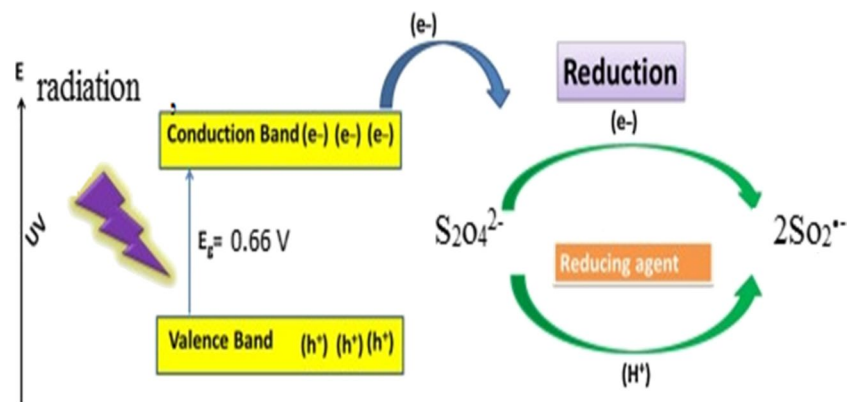
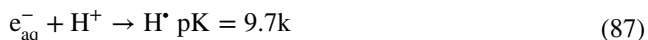


Table 9 Reduced pollutants by UV/dithionite along with free radicals

Contaminations	Process type	Radical species	Reference
Arsenite	UV/dithionite	$\text{SO}_2^{\bullet-}$ , $\text{S}_2\text{O}_4^{\bullet-}$ , $\text{SO}_3^{\bullet-}$	Jung et al. (2018a)
Acid yellow 17 (AY17)	UV/dithionite	$\text{SO}_2^{\bullet-}$ , $\text{H}^\bullet$ , $\text{SO}_3^{\bullet-}$ , $e_{\text{aq}}^-$	Yazdanbakhsh et al. (2021)
Selenite	UV/dithionite	$\text{S}_2\text{O}_4^{\bullet-}$ , $\text{S}_2\text{O}_3^{\bullet-}$ , $\text{SO}_3^{\bullet-}$ , $\text{S}^{\bullet-}$ , $e_{\text{aq}}^-$	Jung et al. (2018b)



Other chain reactions that lead to radical production are described below:

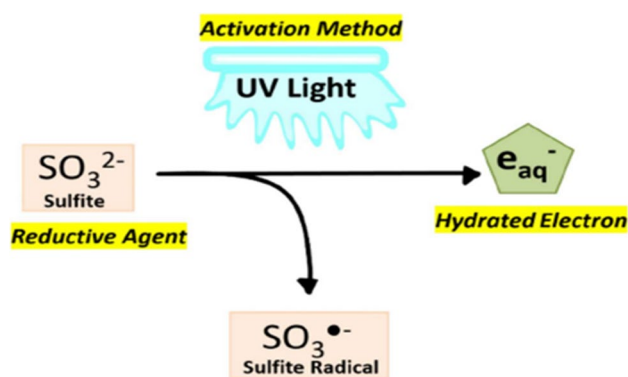


It can be seen that the strong radicals including  $\text{H}^\bullet$  (−2.9 V),  $\text{SO}_3^{\bullet-}$ , and  $e_{\text{aq}}^-$  (−2.3 V) are formed in this process. From Fig. 13, the graphical schematic of RRR production in the UV/sulfite process can be seen. It is also pointed out that compared with common reluctance (i.e., ferrous iron, sulfide, dithionite), the interaction between UV beam and sulfite reluctant is demonstrated to be the highest effective ARP, which corresponds to the massive yield of  $e_{\text{aq}}^-$  therein (Milh et al. 2021). In Table 10, the possible radicals produced and pollutants removed by the UV/sulfite process are explained in detail. One of the most obvious features of the process is that the UV/sulfite process can reduce pollutants in anaerobic conditions when most processes cannot be used in anaerobic conditions. Also, in cases where the use of  $\text{H}_2\text{O}_2$  is not preferred, the UV/ $\text{SO}_3^{2-}$  process may be an interesting alternative. However, it should be noted that the reaction rate constants of the UV/ $\text{SO}_3^{2-}$  process in general are lower than those of a UV/ $\text{H}_2\text{O}_2$  process (Liu et al. 2014; Xiao et al. 2017; Xie et al. 2017).

On the other hand, studies showed that the UV/sulfite process produced more diverse free radicals in aerobic conditions than in anaerobic conditions. Therefore, the presence of oxygen improves the ability of the process to eliminate reducible pollutants (Cao et al. 2021). UV/sulfite has been used to eliminate various contaminations, some of which are mentioned in Table 10, and the possible radicals produced during each process are as well mentioned.

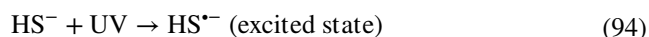
### UV/sulfide

Another sulfur-containing compound used in reduction processes to produce reduced radicals is sulfide ( $\text{S}^{2-}$ ). Sulfide



**Fig. 13** The graphical schematic of RRR production in the UV/sulfite process

can absorb UV beam at 190–250 nm, and hydrogen sulfide ( $\text{H}_2\text{S}$ ) compounds are used to supply sulfide ions. In Eq. 94, producing  $\text{HS}^{\bullet-}$  radical in the UV/sulfide process is mentioned, but the study regarding the use of the UV/sulfide process is limited.



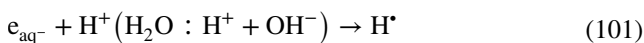
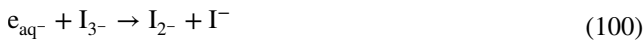
### UV/iodide

Another mineral compound that can be used as a reducer agent to produce reduced radicals in ARP is iodide ( $\text{I}_2$ ). Iodide can be excited by UV beams because iodide can absorb UV beams at 248 to 254 nm. Therefore, this property leads to the occurrence of an ARP between UV and iodide, and a large number of  $e_{\text{aq}}^-$  reducer radicals in the aqueous solutions can be created. In other words, the Van der Waals bond between two atoms of the iodine element is broken by the absorption of ultraviolet rays at 254 nm, and the result is creating iodine radicals. The graphic schematic of the UV/iodide process to produce free reducer radicals is depicted in Fig. 14. Studies have shown that the UV/iodide system produces fewer free radicals than the UV/sulfite system, and its possible cause is attributed to the presence of scavengers. In the UV/iodide system, the reducing agent is iodine ions, and in order to prepare them, potassium iodide is used. The simultaneous presence of UV beam and iodide ions in an aqueous solution leads to the production of reducing radicals during a chain of reactions, which is mentioned below (Eq. 95):



**Table 10** Reduced contaminants by UV/sulfite along with free radicals

Contaminations	Process type	Radical species	Reference
Hexafluoropropylene oxide dimer acid	UV/sulfite	$e_{aq}^-$ , $H^\bullet$	Bao et al. (2018)
Trichlorophenol	UV/sulfite	$SO_3^{\bullet-}$ , $e_{aq}^-$ , $H^\bullet$	Sheikhmohammadi et al. (2019)
Trichloroethylene	UV/sulfite	$SO_3^{\bullet-}$ , $e_{aq}^-$ , $H^\bullet$	Jung et al. (2015)
1,2-Dichloroethane	UV/sulfite	$SO_3^{\bullet-}$ , $e_{aq}^-$ , $H^\bullet$ , $SO_2^{\bullet-}$ , $HS^\bullet$	Liu et al. (2014)
CCl4	UV/sulfite	$SO_3^{\bullet-}$ , $e_{aq}^-$ , $H^\bullet$ , $O_2^{\bullet-}$	Wang et al. (2021b)
Atrazine	UV/sulfite	$SO_3^{\bullet-}$ , $e_{aq}^-$ , $H^\bullet$	Tian et al. (2017)
4-Bromophenol	UV/sulfite	$SO_3^{\bullet-}$ , $e_{aq}^-$	Xie et al. (2017)
Bromate	UV/sulfite	$SO_3^{\bullet-}$ , $e_{aq}^-$	Xiao et al. (2017)

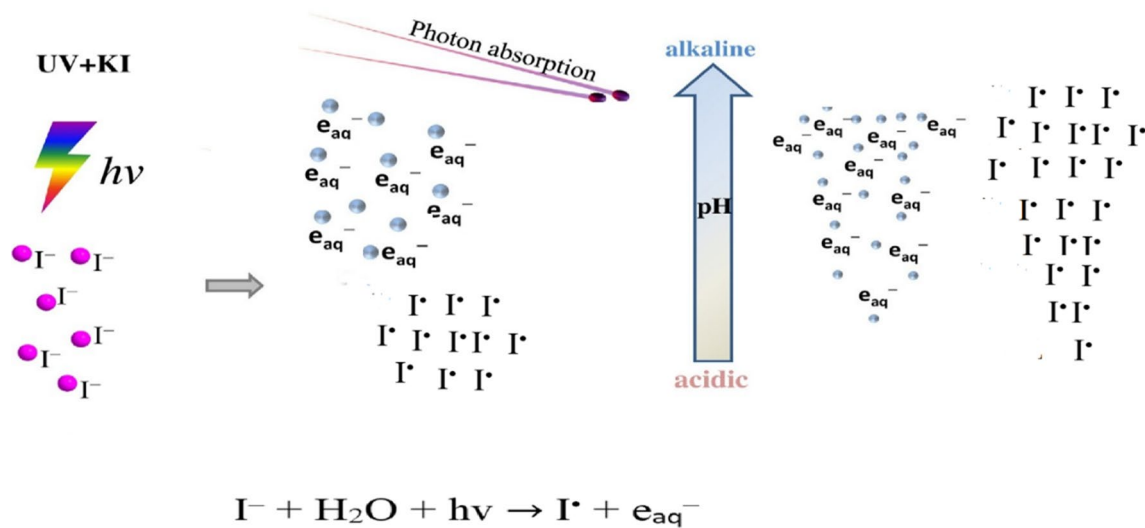


UV/iodide process has been used both alone and in combination with the AOP process to remove various pollutants, including N-nitrosodimethylamine, perfluorooctane sulfonate, Ocuflox, and codeine phosphate, which is mentioned in Table 11, and the corresponding radical for each process is indicated as well. Iodide has a

higher molar absorption coefficient at 254 nm than sulfite; this means that the ability to produce hydrated electrons by the process of UV/iodide is greater than that of the UV/sulfite system. However, some studies have indicated that target contaminant transformation rates are faster in the UV/sulfite system compared to the UV/iodide system. On the other hand, iodine ion has high electronegativity properties, and depending on the conditions of the aqueous solution, it can have a negative or positive effect on the UV/iodide system.

### UV/ferrous

Ferrous iron ( $Fe^{2+}$ ) is one of the parameters that can be used in both oxidation and reduction. The Fenton process, the simplest AOP, will occur when ferrous iron is simultaneously homogenized in an aqueous media with hydrogen peroxide ( $H_2O_2$ ), where ferrous iron plays the role of catalyst, and by activating  $H_2O_2$ , hydroxyl radical ( $OH^\bullet$ ) is produced. Moreover, when ultraviolet rays are irradiated to an aqueous



**Fig. 14** The graphical schematic of RRR production in the UV/iodide system



**Table 11** Reduced contaminants by UV/sulfite along with free radicals

Contaminations	Process type	Radical species	Reference
N-nitroso dimethylamine	UV/iodide	$I^\bullet, e_{aq}^-, H^\bullet$	Sun et al. (2017b)
Perfluorooctane sulfonate	UV/iodide	$I^\bullet, e_{aq}^-, H^\bullet, HA^{+\bullet}$	Sun et al. (2017a)
Ocuflox	AOP + ARP (UV/iodide)	$I^\bullet, e_{aq}^-, OH^\bullet$	Rasoulzadeh et al. (2022)
Codeine phosphate	AOP + ARP (UV/iodide)	$I^\bullet, e_{aq}^-, H^\bullet, OH^\bullet$	Azizi et al. (2021)

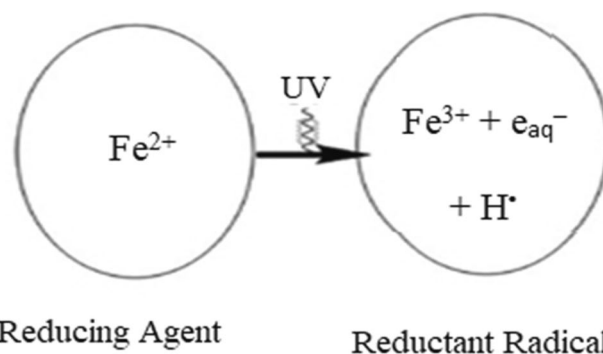
bed containing ferrous iron in the presence of oxygen, the result is an AOP and the production of  $OH^\bullet$ . However, an ARP occurs and a reducing radical ( $e_{aq}^-$ ) is created when UV beams are irradiated to  $Fe^{2+}$  ions in the absence of oxygen in a water media. The solution of  $Fe^{2+}$  absorbs the UV beam with a maximum of 220 nm, and UV irradiation promotes the formation of hydrogen and aqueous electrons. The overall reaction of the UV/ $Fe^{2+}$  process is given in Eq. 103.



Hydrogen and aqueous electrons are the most likely reductant agents that are formed in the UV/ $Fe^{2+}$  process. The graphical schematic of RRR production in the UV/ $Fe^{2+}$  system is seen in Fig. 15. UV/ $Fe^{2+}$  process has rarely been utilized until now as an independent process for free reducer radical generation and removal of a pollutant. For example, in a study, the UV/ $Fe^{2+}$  process was utilized to eliminate vinyl chloride, and the only active species produced was  $e_{aq}^-$  (Liu et al. 2013). In other words, the UV/ $Fe^{2+}$  process has been anaerobically utilized to improve other basic oxidation processes. Of these, UV/US/ $Fe^{2+}$  (bisphenol degradation) (Torres et al. 2007), UV/ $Fe^{2+}$ / $H_2O_2$  (dye degradation) (Kang et al. 1999), UV/ $S_2O_8^{2-}$ / $Fe^{2+}$  (diclofenac sodium removal) (Rehman et al. 2021) processes, etc. can be mentioned. The most important unfavorable feature of the UV/ $Fe^{2+}$  process is the reduction of ferrous iron to ferric iron, which can lead to the formation of sediments and clogging. But in order to prevent the problem, it combines with other processes.

## Conclusion

In this study, the application of ultraviolet rays in redox reactions was reviewed. The results indicated that ultraviolet irradiation is an important parameter in oxidation and reduction reactions so its combination with any oxidizing or reducing agent leads to the production of free radicals. In other words, ultraviolet rays activate oxidizing and reducing substances and improve pollutant detoxification by producing free radicals. The most possible and the strongest radical species in the redox process based on UV irradiation were included reducer ( $I^\bullet, H^\bullet, OH^\bullet, SO_3^{\bullet-}, e_{aq}^-, SO_2^{\bullet-}, HS^\bullet, S_2O_4^{\bullet-}, S_2O_3^{\bullet-},$  and  $S^{\bullet-}$ ) and oxidizer ( $OH^\bullet, O_2^{\bullet-},$



**Fig. 15** The graphical schematic of RRR production in the UV/ $Fe^{2+}$  system

$O_3^{\bullet-}, HO_2^\bullet, HO_3^\bullet, HO_4^\bullet, SO_5^{\bullet-}, SO_4^{\bullet-}, O^\bullet,$  and  $S_2O_8^{\bullet-}$ ) agents. Therefore, it can be concluded that the UV ray is an inseparable factor in redox processes and improves the processes directly and indirectly.

**Author contribution** All persons who meet authorship criteria are listed as authors along with related activities. Kindly, you can see the role of each author below:

MTS: presenting research ideas, conception and design of study, and acquisition of data.

AAE: revising the manuscript and improving English.

AHP: drafting the manuscript, revising the manuscript, and improving the English.

AR: analysis and/or interpretation of data, drafting the manuscript, and acquisition of data.

KK and HA: conception and design of study and acquisition of data.

## Declarations

**Ethics approval** This work received ethical approval from Hamadan University of Medical Sciences (IR.UMSHA.REC.1401.315).

**Competing interests** The authors declare no competing interests.

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