# Degradation of bisphenol S – a contaminant of emerging concern - by synergistic ozone and percarbonate based AOP

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## ARTICLE INFO

## ABSTRACT

Degradation of bisphenol S was studied using ozone activated by sodium percarbonate and the effectiveness of optimized process was compared with the peroxone process. The influence of several factors including sodium percarbonate concentration, ozone dose, pH, and water matrix were investigated. A synergetic coefficient of 3.84 was achieved for the combination of sodium percarbonate and ozone, confirming the effectiveness of this hybrid process. Scavenging tests revealed, that carbonate radicals, hydroxyl radicals, superoxide radicals, and singlet oxygen contributed to the degradation of bisphenol S. At the same operating condition, degradation effectiveness values of 99% and 81% were obtained by ozone combined with sodium percarbonate and hydrogen peroxide, respectively, demonstrating the superiority of sodium percarbonate over hydrogen peroxide in combination with ozone for the degradation of bisphenol S. Low concentration of inorganic anions had a negligible effect on the degradation, while carbonate ions increased the first-order degradation rate constant by 56%.

## 1. Introduction

Emerging contaminants constituting a large quantity of new unnaturally occurring substances in water have known or suspected detrimental effects on living organisms [1–3]. These pollutants consist of pharmaceutical, surfactants, personal care products, industrial additives and solvents, household products, and pesticides [4,5]. In this group of pollutants, synthetic organic compounds known as endocrine-disrupting chemicals (EDCs) can be regarded as a serious threat to sustainability of many creatures since they cause numerous adverse impacts on animal health, especially on their reproduction and development system [6–8]. Plasticizers are

one of the dangerous chemicals included in the category of EDCs. Bisphenol (BP) is one of the common chemicals used as plasticizers, which have different categories such as A, B, S, F, AF, etc., and among them BPA were widely used in the plastic production and had many applications [9–11]. These compounds are easily leachable and contaminate the aquatic environment and pose environmental hazards. BPA can be regarded as a weak estrogen receptor agonist and can cause harmful influence, especially on reproduction and development system of human beings [12,13]. As a result, many countries banned the use of BPA in the industrial application. As an alternative to BPA, industries started extensively using their structural analogues, especially BPS. The emissions of BPS and its concentration in the environment probably will increase as its production and use is extremely growing. It is utilized as an additive in pesticides, colorfast agents, flame retardants, dyestuffs, baby bottles, food packaging material, etc. [14,15]. Thus, studies on degradation of this pollutant in aqueous conditions fall into the technology oriented studies on wastewater treatment processes. However, studies have indicated that BPS can cause the same genotoxicity and estrogenic activity as BPA, intensify lymphocyte proliferation, be problematic for physiologic function in both human beings and rodents, and even be more environmentally persistent than BPA [16, 17]. BPS is more thermally stable and possesses higher adsorption and permeability towards skin. Hence, the release of effluent from a variety of industries such food industry, containing BPS, into the environment can have high hazardous effects [15,18]. A considerable amount of BPS usually passes through secondary treatment procedures and subsequently, enters the aquatic environment [19]. The incorporation of sulfonic group in the molecular structure of BPS causes its resistance to biodegradation, therefore, the biodegradation of this substance is not usually efficient, and it is required to develop more efficient method for the removal of this pollutant [20,21]. Therefore, a proper treatment technology must be addressed and considered for BPS as well.

Advanced oxidation processes (AOPs) are one of the efficient techniques utilized for the degradation of organic pollutants including plasticizers [22–25]. In this process, reactive oxidizing species are generated in situ, which can react with the organics at high rate. Various AOPs attempted for the plasticizers degradation are Fenton processes [26], cavitation-based processes [27,28], persulfate-based processes [29], ozonation [30], and photocatalytic oxidation [31]. An attracting feature of AOPs is the transformation and even mineralization of many organic contaminants which are resistant to biological methods and leads to a reduction in their potential danger to the environment and human life [32,33]. There are several studies detailing the degradation of BPA, but the degradation of BPS has been rarely investigated. Frankowski et al. [34] have studied the degradation of BPS using photo-Fenton and reported 100% degradation effectiveness at optimum conditions during 10–20 min. Mehrabani-Zeinabad et al. [35] have reported that a BPS degradation effectiveness above 95% was achieved using ultraviolet combined with ozone. Cao et al. [36] have investigated the effect of a variety of factors on the photodegradation of BPS and reported 100% degradation effectiveness at optimized conditions. Wang et al. [37] have reported almost 100% degradation effectiveness of BPS by heat-activated persulfate at a temperature of 70 °C in 120 min.

Ozone ($O_3$) is known as a powerful oxidant which has various applications such as disinfection, decolorization, removal of odor, and sludge reduction [38]. It is highly selective towards specific functional groups like unsaturated bonds and deprotonated amins [39]. The oxidation in $O_3$-based processes takes place through two key oxidation pathways. These pathways are the direct oxidation by $O_3$ molecules and indirect oxidation performed by generated $^\bullet OH$ [40,41]. Although $O_3$ possesses a high redox potential, oxidation through the direct oxidation by sole $O_3$ usually leads to a low degradation efficiency of organic pollutants [42]. Extensive studies have indicated that the oxidation of a variety of compounds by $^\bullet OH$ has greater reaction rate and mineralization efficiency compared to oxidation by $O_3$ molecules [43]. Some researchers have regarded the combination of $O_3$ and hydrogen peroxide ($H_2O_2$), called peroxone process, as a solution for this problem since it can result in the generation of more hydroxyl radicals ($^\bullet OH$) [44–46]. According to a sustainability viewpoint, a method is required to be efficient, environmentally-friendly, and easy to be employed to be able to be transferred from lab scales to industrial applications, and concentrated $H_2O_2$ has the drawbacks of difficult storage and transportation [43].

Sodium percarbonate (SPC) with formula $Na_2CO_3 \bullet 1.5H_2O_2$ can be considered as solid carrier of $H_2O_2$ [47]. One of the superiorities of percarbonate (PC) over $H_2O_2$ are safety in transportation and storage without any risk of spilling, splashing, and corrosion. It costs almost 3 times less than $H_2O_2$, which can make it more suitable for industrial-scale applications. The controlled release of $H_2O_2$ can be another advantage of this oxidant [48,49]. Therefore, this safety and convenience has made PC an attractive oxidant employed in wastewater treatment [50]. In addition, it is more environmentally-friendly and safer as its toxicity is lower compared to $H_2O_2$ [51]. Moreover, compared to some other AOPs like Fenton reagent, PC-based AOPs can be applied in a wide range of pH [52]. Although the low degradation efficiency and mineralization degree induced by direct PC can be regarded as a negative point, after its activation, the generated reactive species can contribute to the degradation of pollutants and result in high efficiency [53]. PC-based treatment processes can be applied for the degradation of a variety of contaminants. These combined processes have been reported to be effective for the treatment of pesticide-laden wastewater [54], dye industry effluent [55], and degradation of chlorinated organic compounds [56], antibiotics [57], aniline [58], etc. A variety of factors such as the concentrations of oxidants, anions, pH, etc. can positively or negatively influence the effectiveness of PC-based ozonation [59]. An increase in pH with the addition of SPC is an advantage in the ozonation process [60].

By considering all the above facts, the aim of the current investigation was to (1) study the degradation of BPS using a combination of SPC and $O_3$ (SPC–$O_3$) (2) compare the degradation effectiveness with peroxone process (3) investigate a variety of parameters such as the concentration of oxidants and initial pH of solution, (4) identify the major species formed in the medium, (5) and study the practicability of the processes in the presence of co-existence of inorganic anions.

## 2. Materials and methods

### 2.1. Chemicals

Sodium percarbonate ($Na_2CO_3 \bullet 1.5H_2O_2$) was provided from Sigma-Aldrich (USA). Bisphenol S (97%) was purchased from AmBeed (USA). Sodium hydroxide (99%), acetonitrile, methanol, phenol, sodium nitrate (98%), potassium iodide, thiosulfate, and hydrogen peroxide (30%) were purchased from POCH (Poland). Sodium carbonate, sodium sulfate (99%), and sulfuric acid (95%) were provided from Chempur (Poland). *Para*-benzoquinone (99%) was purchased from Acros Organics (China). Sodium chloride was purchased from Stanlab (Poland) and 1,4-Diazabicyclo [2.2.2]octane (DABCO) was purchased from Chemat (Poland). All materials were of analytical grade and were utilized without further purification. $O_3$ was generated by a Tytan 32 ozone generator (Erem).

### 2.2. Procedure

Aqueous solution of BPS was prepared using a magnetic stirrer (700 rpm) at the room temperature. SPC was added to the solution at the beginning of experiments. pH was measured by a pH test paper (Chemland, Poland). Experiments were performed in a batch reactor and a reaction temperature of $20 \pm 2\ °C$ was maintained by circulating cooling water through a cooling coil installed into the reactor. The total treatment time for all experiments was 60 min. All experiments were conducted in deionized water at an initial BPS concentration of 50 ppm. $O_3$ was continuously injected from the ozone generator to the reactor through a nozzle throughout the treatment. Initial samples were taken before the start of the treatment and then samples were taken every 10 min until 60 min.

To identify the reactive species, a selected scavenger with a ratio of 30:1 (scavenger: SPC) was used. The effect of inorganic anions was studied with the addition of an inorganic anion at a ratio of 100:1 and 1000:1 (anion: BPS). The degradation of BPS with SPC-$O_3$ system was performed initially. The degradation effectiveness was determined by Eq. (1). First-order degradation rate constant was also calculated using Eq. (2).

$$Degradation\ effectiveness\% = \frac{C_0 - C}{C_0} \times 100 \tag{1}$$

$$k = \frac{ln\left(\frac{C_0}{C}\right)}{t} \tag{2}$$

where k, $C_0$, t, and C refer to first-order degradation rate constant, the initial concentration of BPS, the time of reaction (min), and the concentration in time t, respectively.

### 2.3. Investigation of ozone emission from the reactor

The emission of unreacted $O_3$ was determined by potassium iodide wet-chemistry test [61]. A piece of PTEE tube was attached to a
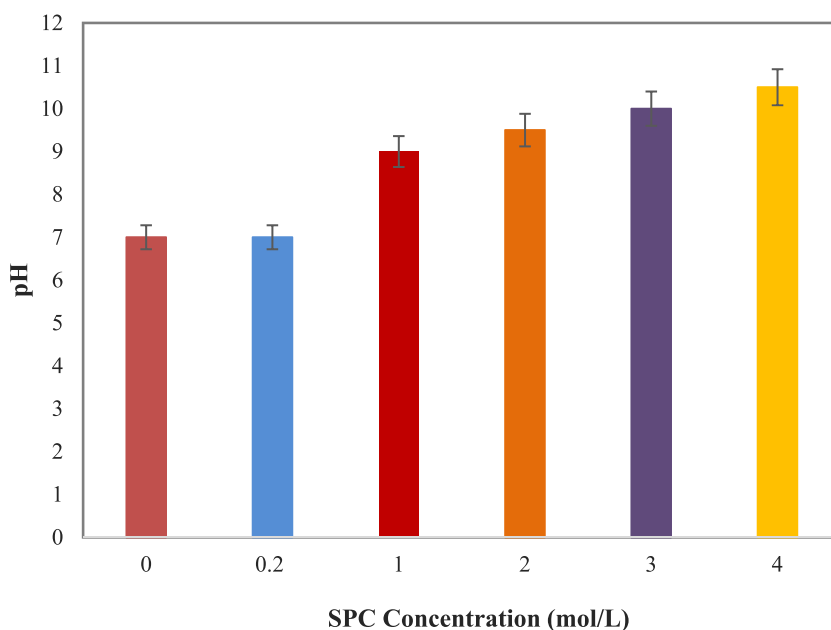
**Fig. 1.** Changes in the initial pH of solution through the addition of SPC (BPS concentration = 50 ppm).

dedicated outlet of the reactor. The other end of the tube was connected to two scrubbers which contained 400 mL of potassium iodide aqueous solution (2%), each. The unreacted $O_3$ was absorbed in the scrubbers. Then, the solution in both scrubbers was titrated with $Na_2S_2O_3$ (0.01 N) to determine how much $O_3$ was emitted from the reactor.

### 2.4. Analysis

The concentration of BPS in the sample was analyzed using high performance liquid chromatography (HPLC) equipped with a C18 column which had a dimension of 4.6 × 150 mm obtained from Agilent Technologies (USA). The mobile phase applied in the determination was a mixture of acetonitrile: water (30:70 v/v) under isocratic conditions with a flow rate of 0.8 mL min$^{-1}$. The detection wavelength was 260 nm. All the determinations were performed twice, and the average values of two analyses were reported.

## 3. Result and discussion

### 3.1. Degradation study of BPS by SPC-O$_3$ system

Combined application of SPC and $O_3$ (SPC–$O_3$) can result in the formation of reactive radicals such as hydroxyl radicals ($^\bullet$OH), superoxide radicals ($O_2^{\bullet-}$), and carbonate radicals ($CO_3^{\bullet-}$), which can attack pollutant molecules and degrade them into less harmful compounds [59]. The degradation of BPS by SPS-$O_3$ is attributed to several possible reactions between both chemicals as well as the fact that SPC can induce alkaline conditions in the solution. The changes made in the initial pH of solution as a result of the addition of different concentrations of SPC are shown in Fig. 1.

The alkaline condition can be beneficial for the decomposition of $O_3$ and generation of reactive $^\bullet$OH, which can happen through three following paths initiated by hydroxyl ions (Eqs. (3)–(11)) [29].

Path 1:

$$O_3 + OH^- \rightarrow HO_2^\bullet + O_2^{\bullet-} \tag{3}$$

$$HO_2^\bullet \leftrightarrow H^+ + O_2^{\bullet-} \tag{4}$$

$$O_3 + O_2^{\bullet-} \rightarrow O_3^{\bullet-} + O_2 \tag{5}$$

$$O_3^{\bullet-} + H_2O \rightarrow {}^\bullet OH + OH^- + O_2 \tag{6}$$

Path 2:

$$O_3 + OH^- \rightarrow HO_2^- + O_2 \tag{7}$$

$$O_3 + HO_2^- \rightarrow {}^\bullet OH + O_2^{\bullet-} + O_2 \tag{8}$$

Followed by Eq. (5) and Eq. (6).

Path 3:

$$O_3 + OH^- \rightarrow HO_2^- + O_2 \tag{9}$$

$$O_3 + HO_2^- \rightarrow O_3^{\bullet-} + HO_2^\bullet \tag{10}$$

$$HO_2^\bullet + OH^- \leftrightarrow O_2^{\bullet-} + H_2O \tag{11}$$

Followed by Eq. (5) and Eq. (6).

In addition, $H_2O_2$ originated from SPC can accelerate the decomposition of $O_3$ and consequently, the generation of $^\bullet$OH according to the Eqs. (12) and (13) [60].

$$2Na_2CO_3 \bullet 3H_2O_2 \rightarrow 2Na_2CO_3 + 3H_2O_2 \tag{12}$$

$$O_3 + H_2O_2 \rightarrow 2^\bullet OH + 3O_2 \tag{13}$$

Furthermore, the induced alkaline condition shifts Eq. (14) towards the production of more $HO_2^-$, which can lead to the production of more $^\bullet$OH according to Eq. (8) and Eq. (10) [42].

$$H_2O_2 \leftrightarrow HO_2^- + H^+ \tag{14}$$

Carbonate ions can be regarded as a scavenger for reactive $^\bullet$OH according Eq. (15). However, generated $CO_3^{\bullet-}$ can selectively oxidize electron-rich compounds like BPS and contribute to the degradation of the pollutant.

$$CO_3^{2-} + {}^\bullet OH \rightarrow CO_3^{\bullet-} + OH^- \tag{15}$$

Singlet oxygen ($^1O_2$) is a relatively strong oxidant which can be produced through the reaction of $^\bullet$OH or $HO_2^\bullet$ with $O_2^{\bullet-}$ according to Eq. (16) and Eq. (17) [62,63]. Therefore, it can also have an effect on the degradation of pollutants using $O_3$-based processes.

$$\bullet O_2^- + \bullet OH \rightarrow {}^1O_2 + OH^- \tag{16}$$

$$\bullet O_2^- + HO_2^\bullet + H^+ \rightarrow {}^1O_2 + H_2O_2 \tag{17}$$

Although SPC acts as a base increasing the pH of water, $H_2O_2$ has acidic property decreasing the pH of solution [64]. Since alkaline pH is the desired condition in $O_3$-based processes, the adjustment of pH in $H_2O_2$–$O_3$ requires higher cost compared to SPC-$O_3$.

### 3.1.1. Comparison of BPS degradation effectiveness by SPC-$O_3$ and SPC-$H_2O_2$

In order to determine the superiority of SPC over $H_2O_2$ for the degradation of BPS by $O_3$, exclusively by increasing pH, experiments were performed using sole $O_3$, SPC- $O_3$, and $H_2O_2$–$O_3$ at a constant pH of 11 adjusted by NaOH, an $O_3$ flow rate of 0.019 mol h$^{-1}$, a SPC concentration of 1 mol L$^{-1}$ or a $H_2O_2$ concentration of 1.5 mol L$^{-1}$. The degradation effectiveness and kinetic results are illustrated in Fig. 2.

Based on the outcomes, SPC-$O_3$ resulted in a maximum degradation effectiveness of 99% at a first-order degradation rate constant of 0.0755 min$^{-1}$, while sole $O_3$ led to a degradation effectiveness of 92% at a first-order degradation rate constant of 0.0478 min$^{-1}$ and $H_2O_2$–$O_3$ caused a degradation effectiveness of 81% at a first-order degradation rate constant of 0.0323 min$^{-1}$. Thus, it is obvious that the SPC-$O_3$ is the best solution, even despite its positive effect on increasing pH, among these three AOPs for the degradation of BPS in aqueous medium. In $H_2O_2$–$O_3$, the addition of $H_2O_2$ all in once to the solution can increase the degradation effectiveness at the beginning of the experiment but after a while its scavenging effect can lead to a reduction in the concentration of available $\bullet OH$ required for the degradation of the pollutant, while in SPC-$O_3$, the slow release of $H_2O_2$ from SPC keeps a sustained supply of $H_2O_2$ in the solution [65]. The enhancement in the degradation effectiveness made as a result of the presence of SPC can be related to the mild release of $H_2O_2$, which can suppress its scavenging effect and it can react with $O_3$ and produce reactive $\bullet OH$. Furthermore, PC can virtually act as a buffer and prevent from a considerable reduction in the pH of solutions throughout the experiment [64].

The changes in the pH of solution during the experiments are demonstrated in Fig. 3. As it is observed, the decrease in the solution pH in SPC- $O_3$ is smoother than sole $O_3$.

The generation of $CO_3^{\bullet-}$ might be another reason for the promotion of the degradation of BPS using SPC-$O_3$.

### 3.1.2. Identification of radical species

Several species such as hydroxyl radical ($\bullet OH$), superoxide radical ($O_2^{\bullet-}$), and carbonate radical ($CO_3^{\bullet-}$) could take part in the degradation of BPS in SPC-$O_3$. Radical quenchers are capable to react with a variety of oxidative species but their selectivity is towards a particular radical.

Radical quenching experiments were performed to identify the reactive species which participated in the degradation of this pollutant at a pH of 9 and a rox ($\frac{Mole_{(SPC+Ozone)}}{Mole_{BPS}}$) of 5.02. The scavengers were used in a high mole ratio of 30:1 ($\frac{Scavenger}{SPC} = 30$) to ensure the complete scavenging of radicals. The degradation effectiveness and kinetic results are demonstrated in Fig. 4.

Methanol is known as a scavenger for $\bullet OH$, reacting with it at a second-order reaction rate constant of $9.7 \times 10^8$ M$^{-1}$ s$^{-1}$ and slowly reacts with other radicals [27]. Since $\bullet OH$ usually involves in the degradation of organic compounds using $O_3$-based processes, methanol was employed to study the involvement of this radical in the degradation of BPS. Carbonate anion can react with $HO^\bullet$ and produce $CO_3^{\bullet-}$ according to Eq. (15) at a second-order reaction rate constant of $3.9 \times 10^8$ M$^{-1}$ s$^{-1}$ [44]. Therefore, phenol was utilized
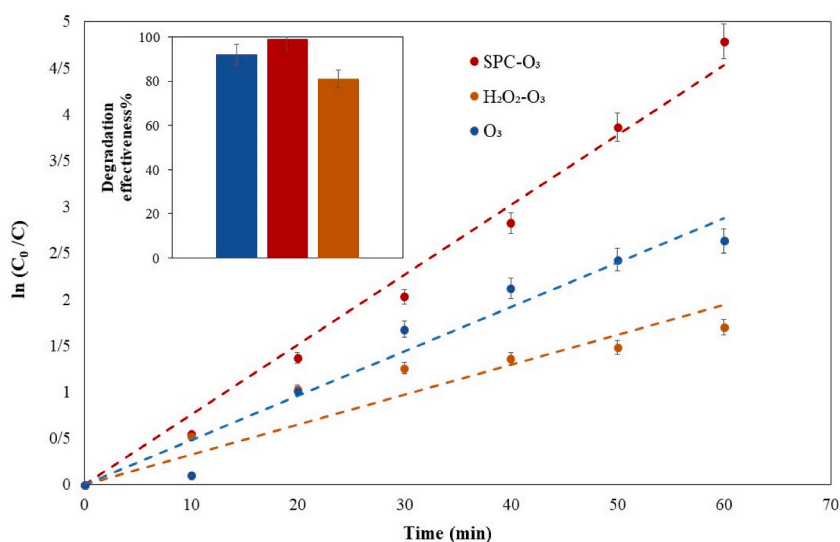


**Fig. 2.** Degradation effectiveness of BPS and kinetic results by $O_3$, $H_2O_2$–$O_3$, and SPC-$O_3$ (pH = 11, $O_3$ flow rate = 0.019 mol h$^{-1}$, SPC concentration = 1 mol L$^{-1}$, $H_2O_2$ concentration = 1.5 mol L$^{-1}$, BPS concentration = 50 ppm).
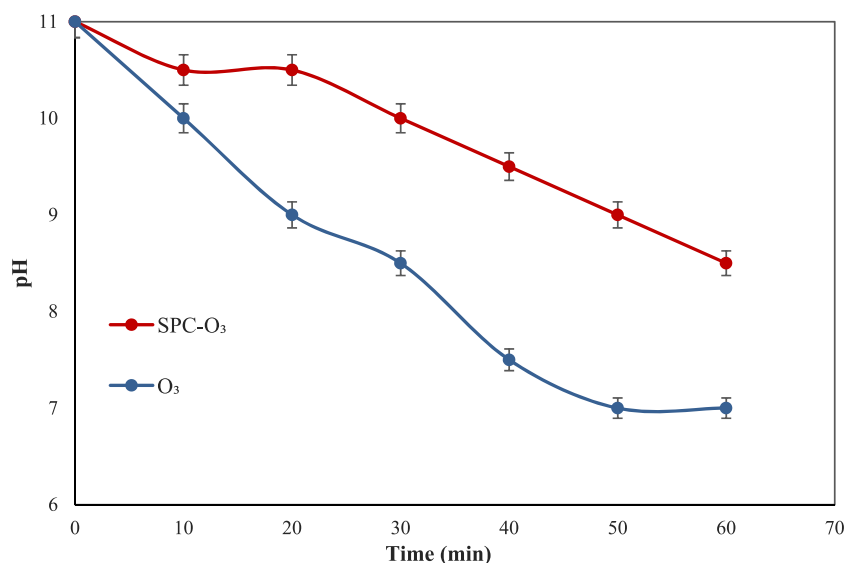
**Fig. 3.** Changes in the pH of solution in $O_3$ and SPC-$O_3$ throughout the experiments ($O_3$ flow rate = 0.019 mol h$^{-1}$, SPC concentration = 1 mol L$^{-1}$, BPS concentration = 50 ppm).



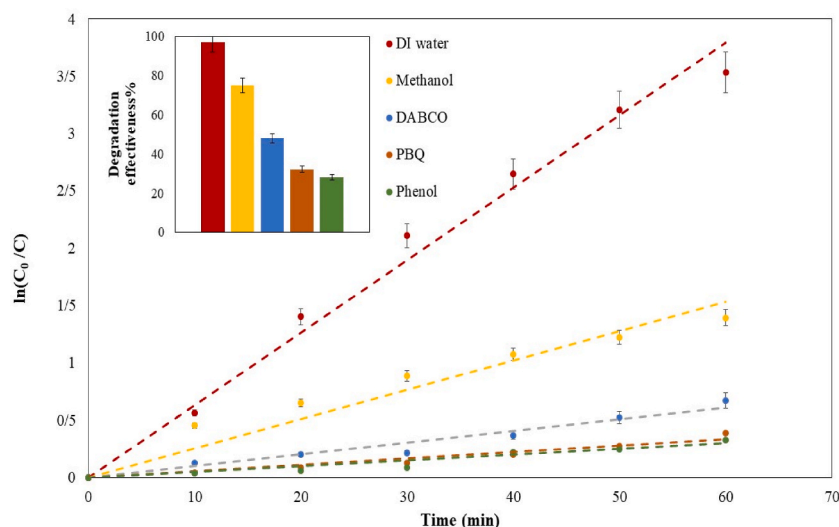**Fig. 4.** Effect of scavengers on BPS degradation effectiveness (pH = 9, BPS concentration = 50 ppm, rox = 5.02, $\frac{Scavenger}{BPS}$ = 5).

to evaluate the participation of $CO_3^{\bullet-}$. Phenol can be regarded as a quencher for both $HO^{\bullet}$ and $CO_3^{\bullet-}$ as it reacts with these radicals at high second-order reaction rate constants of $6 \times 10^8$ M$^{-1}$ s$^{-1}$ and $1.2 \times 10^9$ M$^{-1}$ s$^{-1}$, respectively [50,66,67]. Therefore, a comparison between the results achieved through the presence of methanol and phenol can show the contribution of $CO_3^{\bullet-}$. *Para*-benzoquinone (PBQ) can be used as a quenching agent for $O_2^{\bullet-}$ since it reacts rapidly with this radical at a high second-order reaction rate constants of $(0.9–1) \times 10^9$ M$^{-1}$ s$^{-1}$ [27,68]. DABCO can scavenge $^{\bullet}OH$ and $^1O_2$ at second-order reaction rate constant values of $1.25 \times 10^9$ M$^{-1}$ s$^{-1}$ and $3.1 \times 10^8$ M$^{-1}$ s$^{-1}$, respectively. Hence the difference between the degradation effectiveness achieved in the presence of methanol and DABCO can prove the role of $^1O_2$ in the degradation of BPS using SPC-$O_3$ [69].

According to the results, methanol decreased the degradation effectiveness and first-order degradation rate constant of the process from 97% to 0.0632 min$^{-1}$ to 75% and 0.0255 min$^{-1}$, respectively, proving the generation and moderate contribution of $^{\bullet}OH$ in the degradation of BPS using SPC-$O_3$. The presence of phenol in the solution decreased the degradation effectiveness and first-order degradation rate constant to 28% and 0.005 min$^{-1}$, respectively. The noticeable difference in the degradation efficiencies between methanol and phenol reveals the significant role of $CO_3^{\bullet-}$ as it can selectively oxidize electron-rich substances such as BPS. As it is demonstrated, the addition of p-BQ to the solution caused a drop in the degradation effectiveness to 32%, demonstrating the significance of superoxide radical ($O_2^{\bullet-}$) in the degradation of the pollutant. The presence of DABCO in the solution decreased the

degradation effectiveness and first-order degradation rate constant to 48% and 0.102 $min^{-1}$, respectively. The difference between the degradation effectiveness of BPS in the presence of methanol and DABCO can prove the involvement of singlet oxygen ($^1O_2$) in the degradation of BPS. It should be considered that although this scavenging test can be performed to prove the generation of radicals and their participation in the degradation of pollutants it cannot be employed for investigating the exact quantification of contribution of reactive species as it can be overlaid with this fact that scavenging some radicals prohibits the formation of some other radicals [67].

### 3.2. Effect of SPC concentration

A variety of factors can influence the degradation effectiveness of pollutants using AOPs, and the concentration of oxidants is one of those key factors. To investigate the effect of this parameter, the degradation of BPS was performed using different concentrations of SPC at a constant $O_3$ flow rate of 0.019 mol $h^{-1}$. Since alkaline pH induced by SPC is one of advantages in SPC- $O_3$, pH was not adjusted in these experiments. The degradation effectiveness and kinetics of these experiments are demonstrated in Fig. 5.

As it is observed in Fig. 5, the combination of $O_3$ with SPC enhanced the degradation effectiveness of the pollutant, exhibiting the synergistic effect of them. Based on the results, sole $O_3$ led to a degradation effectiveness of 61% at a first-order degradation rate constant of 0.0174 $min^{-1}$ in 60 min reaction time. No degradation was observed using sole SPC throughout the experiment. An increase in the concentration of SPC till an optimum amount of 1 mol $L^{-1}$ increased the degradation effectiveness and first-order degradation rate constant to 97% and 0.0669 $min^{-1}$, respectively, and beyond this value, they remained almost constant and then underwent a slight decrease.

The synergetic coefficient (SC) of combination of $O_3$ and SPC was 3.84, which was calculated according to Eq. (18):

$$SC = \frac{k_{(O_3+SPC)}}{k_{O_3} + k_{SPC}} \tag{18}$$

Since SC higher than 1 generally exhibits the synergy between oxidants, a SC of 3.84 indicates a noticeable synergistic effect of combined application of $O_3$ and SPC [60].

The obtained results are consistent with the outcomes reported by Yu et al. [43], who investigated the effect of the concentration of SPC on the degradation of dichlorvos and indicated that an increase in SPC up to an optimum value enhanced the degradation of the pollutant, and further increase had an inhabitation effect. The enhanced degradation effectiveness through an increase in SPC concertation can be attributed to the enhanced generation of reactive species through Eqs. (3)–(11) as a result of induced alkaline pH, intensified generation of $^\bullet OH$ through Eqs. (12) and (13) due to the higher concentration of available $H_2O_2$, and enhanced generation of selective carbonate radicals according to Eq. (15).

The decrease observed beyond the optimum value of SPC is probably caused by the excess amount of SPC that forms excessive $H_2O_2$, which is a scavenger for $^\bullet OH$ and can consume it according to Eq. (19).

$$H_2O_2 + {}^\bullet OH \rightarrow HO_2^\bullet + H_2O \tag{19}$$

Therefore, the available $^\bullet OH$ responsible for the oxidation of the pollutant decreased and caused a drop in the first-order degradation rate constant.
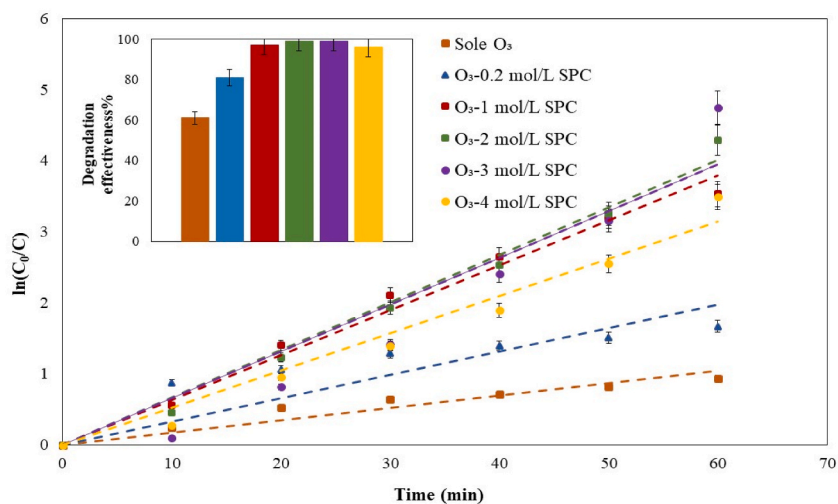


**Fig. 5.** Effect of concentration of SPC on BPS degradation effectiveness ($O_3$ flow rate = 0.019 mol $h^{-1}$, BPS concentration = 50 ppm).

### 3.3. Effect of $O_3$ concentration

The concentration of $O_3$ is another factor impacting the degradation of the pollutant. It is important to assess the optimum value of this oxidant, especially in order to decrease the cost of the decontamination in $O_3$-based process. Hence, the degradation effectiveness of BPC was investigated under 3 different flow rates of $O_3$ at a constant SPC concentration of 1 mol $L^{-1}$ and a constant pH of 11 in 60 min. The degradation effectiveness and kinetic results are demonstrated in Fig. 6.

Analysis of results presented on Fig. 6 confirms that along with an increase in the flow of $O_3$ continuously increased the degradation effectiveness of the pollutant. The utilization of $O_3$ with flow rates of 0.009 mol $h^{-1}$, 0.015 mol $h^{-1}$, and 0.019 mol $h^{-1}$ led to degradation effectiveness values of 16%, 63%, and 99% (k = 0.0755 $min^{-1}$), respectively. This trend was expected and is in agreement with the data reported by Guo et al. [60], who studied the influence of concentration of $O_3$ on the degradation of antibiotic sulfamethoxazole using the same AOP. Obviously, an increase in $O_3$ concentration can enhance its decomposition and the reaction between $O_3$ and $H_2O_2$, and as a result, intensify the formation of $^\bullet OH$ and the degradation of pollutants [70].

### 3.4. Effect of pH

The pH of solutions plays an important role in the degradation of organic pollutants using $O_3$-based processes since it influences the paths of oxidation of pollutants, affecting the effectiveness of decontamination processes [60]. In order to study the influence of pH on the degradation of BPS, the experiments were performed using 4 different pH values of 5, 7, 9, and 11 at a constant rox of 5.02. Due to the fact that $O_3$ is not very effective in acidic conditions, lower pH conditions were not studied. The degradation effectiveness and kinetic outcomes are illustrated in Fig. 7.

It can be seen that an increase in pH enhanced the degradation effectiveness and first-order degradation rate constant, and the pollutants were almost completely degraded (99%) with a first-order degradation rate constant of 0.0755 $min^{-1}$ at a pH value of 11. The degradation effectiveness values of BPS systematically decreased from 97% for pH 9–59% and 50% for pH 7 and 5, respectively. The results, to some extent, can be in accordance to the study performed by Chen and Wang [71], who studied the effect of pH on the degradation of ofloxacin and indicated the agreement of their results with this original view that the degradation of organic compounds using peroxone ($O_3$–$H_2O_2$) is accelerated at alkaline condition. This ascending trend can be attributed to the fact that the decomposition of $O_3$ and consequent generation of $^\bullet OH$ can be facilitated in alkaline condition in which $OH^-$ is utilized as an initiator for the formation of $^\bullet OH$ in this system. In fact, an increase in pH can shift the degradation pathway from direct oxidation by $O_3$ to indirect oxidation performed by reactive radicals, leading to an enhancement in degradation effectiveness. Furthermore, in alkaline condition, more $HO_2^-$ can be generated according to Eq. (14), resulting in the formation of more $^\bullet OH$, and as a result, the intensification of the degradation [40]. It can be again an indication proving that the presence of SPC, as an agent increasing the pH of solution due to hydrolysis of $CO_3^{2-}$ (according to Fig. 1), can result in an enhancement in the degradation of BPS by ozonation, regardless of other positive effects of SPC on the degradation of this pollutant. In addition, SPC is less stable in alkaline condition and can be decomposed faster, contributing to the degradation of pollutants [72].

### 3.5. Effect of inorganic anions

Natural water contains a variety of dissolved compounds such as inorganic anions and other organic substances. As the content of water matrix can extremely influence the effectiveness of employed treatment methods, it is crucial to investigate the impact of
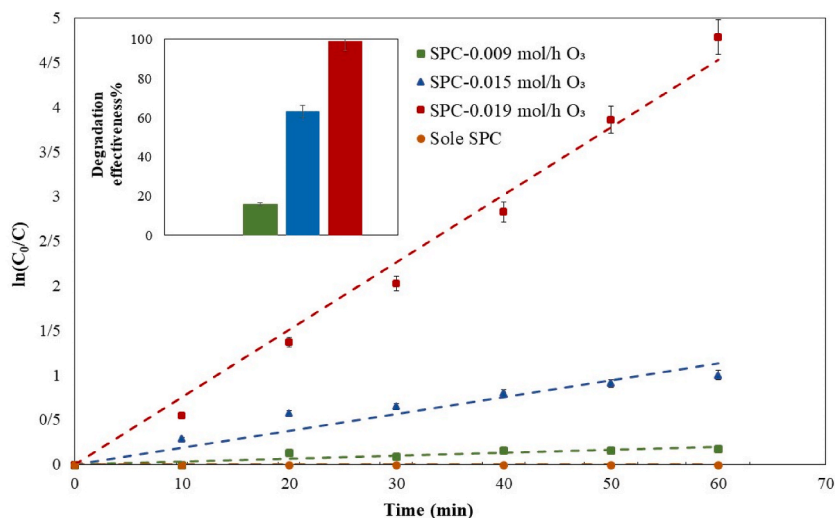


**Fig. 6.** Effect of $O_3$ flow rate on BPS degradation effectiveness (pH = 11, SPC concentration = 1 mol $L^{-1}$, BPS concentration = 50 ppm).
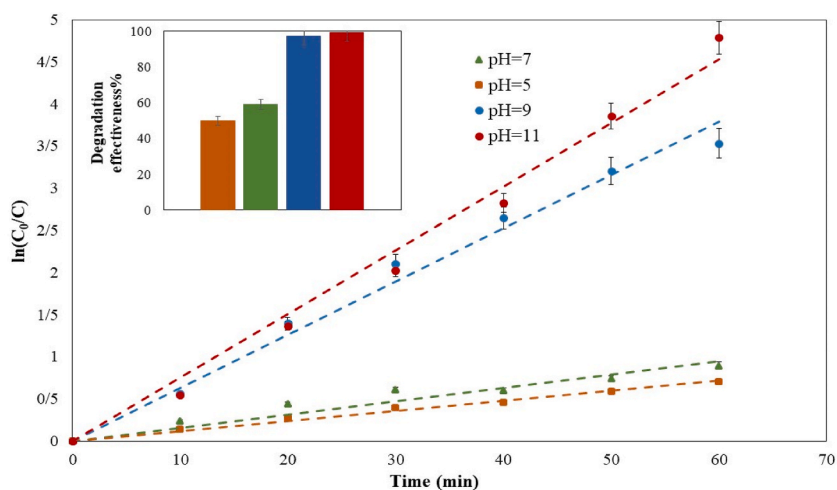
**Fig. 7.** Effect of initial pH of solution on BPS degradation effectiveness (rox = 5.02, BPS concentration = 50 ppm).

common anions existing in natural water on the degradation of pollutants. For this aim, experiments were done at the presence of NaCl, $Na_2SO_4$, $Na_2CO_3$ and $NaNO_3$ at a ratio of 100 ($\frac{Anion}{BPS} = 100$) and a rox of 5.02 in 60 min. The kinetic results of this investigation are demonstrated in Fig. 8.

According to the results, the degradation effectiveness of the pollutant was almost constant at the presence of low concentration of NaCl, $Na_2SO_4$, and $NaNO_3$, however, they negligibly decreased the first-order degradation rate constant. Therefore, it can be concluded that the presence of low concentration of these three anions did not affect significantly the degradation of BPS using SPC-$O_3$. Nevertheless, the promotion effect of the presence of $Na_2CO_3$ was obvious. The addition of $Na_2CO_3$ to the solution led to an increase in the degradation effectiveness of BPS and the complete degradation of this pollutant happened in 50 min at a first-order degradation rate constant of 0.0985 $min^{-1}$. $Na_2CO_3$ does not directly react with $O_3$, but it reacts with $^\bullet OH$ produced by the decomposition of $O_3$ and generates $CO_3^{\bullet}$ [73]. The effect of $CO_3^{2-}$ on $^\bullet OH$-based processes depends on the types of pollutants molecules [74]. The generated $CO_3^{\bullet-}$ can make a contribution to the degradation of organic pollutants and mainly react with electron-rich compounds like (Phenol and compounds containing S and N) with a first-order degradation rate constant in a range from $10^5$ to $10^9$ $M^{-1}$ $s^{-1}$ [27,75]. Hence, in spite of the lower oxidation potential of $CO_3^{\bullet-}$, it can indicate better performance in the degradation of electron-rich compounds, which can be the result of its longer life time and higher selectivity [76–78]. Furthermore, the buffering capacity of $CO_3^{2-}$ prevents a drastic decrease in the pH throughout the process [79]. Hence, it can be another aspect of the positive effect of $CO_3^{2-}$ on the degradation effectiveness of BPS by SPC-$O_3$.

NaCl, $Na_2SO_4$, and $NaNO_3$ were employed at a higher concentration ($\frac{Anion}{BPS} = 1000$) to evaluate the performance of used oxidation system in high salinity conditions and the results are illustrated in Fig. 9.

As it is observed, at a higher concentration, the effect of NaCl was still weak, however, it slightly increased the degradation effectiveness and first-order degradation rate constant to 99% and 0.0744 $min^{-1}$. NaCl can have both inhabitation and promotion effects on the degradation of organic pollutants using AOPs [27]. $Cl^-$ can consume $^\bullet OH$ and produce $Cl^\bullet$ through a series of reactions, and the produced $Cl^\bullet$ can participate in the degradation of pollutants since it has a high oxidation potential of 2.4 V [43]. It can also react with $Cl^-$ and generate $Cl_2^{\bullet-}$, while these reactions are more significant in acidic conditions [73]. The presence of $Cl^-$ can inhibit the degradation of the electron-poor compounds because of the consumption of $^\bullet OH$. Nevertheless, it can enhance the degradation of electron-rich compounds since the generated $Cl_2^{\bullet-}$ can be more selective towards electron-rich centers, resulting in more focused oxidation on target contaminants [27]. Hence, it can be a possible reason for the slight increase in the degradation of BPS in the presence NaCl at its high concentration. $Na_2SO_4$ decreased first-order degradation rate constant to 0.0471 $min^{-1}$. $SO_4^{2-}$ can consume $^\bullet OH$ and generate $SO_4^{\bullet-}$, which possesses a lower oxidation potential compared to $^\bullet OH$ [80]. Also here, more selective system can be obtained. Hence, the slight reduction in the degradation of BPS caused by the addition of $Na_2SO_4$ can be due the scavenging effect of $SO_4^{2-}$ leading to a decrease in the concentration of $^\bullet OH$ [27]. The results show that the impact of high concentration of $NaNO_3$ on the degradation of BPS was much more significant than the effect of $SO_4^{2-}$ and $Cl^{-1}$. The $NO_3^-$ anions decreased the degradation effectiveness and first-order degradation rate constant to 24% and 0.0057 $min^{-1}$. It follows from the fact that $NO_3^-$ is a quencher for reactive $^\bullet OH$ and reduces the amount of $^\bullet OH$ available for the oxidation of pollutants. It reacts with $^\bullet OH$ to form $NO_3^{\bullet}$ which has lower oxidation potential rather than $^\bullet OH$. Based on the results, this radical was unable to oxidize BPS at an appropriate first-order degradation rate constant causing a drastic decrease in the degradation effectiveness. However it should be taken into account that the presence of both $NO_3^-$ and $SO_4^{2-}$ can promote the degradation of some organic pollutants using AOPs [74]. These results are positive in overall, as so high concentration of nitrates are not common for industrial effluents. Nitrites at this concertation could be found only in case of some fertilizer related effluents or in effluents after nitrification process. In second case, it would be more practical to first perform denitrification stage (biological conversion of nitrates to $N_2$) and later apply AOP stage for degradation of BPS.
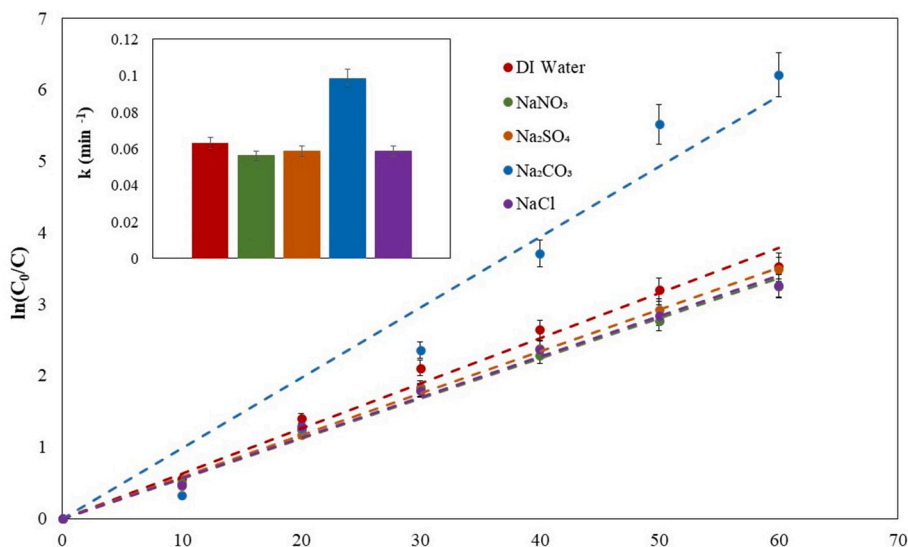
**Fig. 8.** Effect of low concentration of inorganic anions on BPS degradation effectiveness ($\frac{Anion}{BPS} = 100$, **rox = 5.02,** pH = **9, BPS concentration =** **50 ppm).**
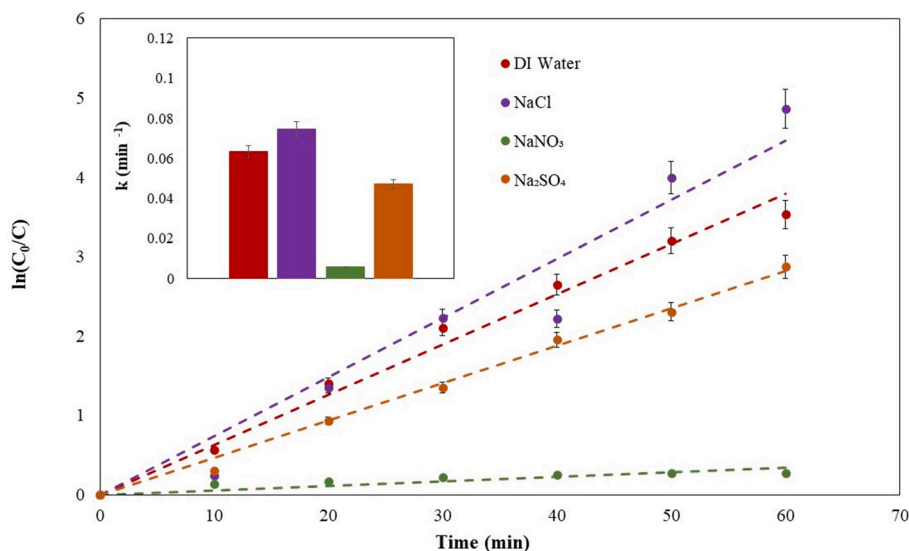


**Fig. 9.** Effect of high concentration of inorganic anions on BPS degradation effectiveness ($\frac{Anion}{BPS} = 1000$, **rox = 5.02,** pH = **9, BPS concentration =** **50 ppm).**

## 4. Conclusion

Degradation of BPS was effectively performed using the combined process of SPC-O$_3$. A degradation effectiveness of 99% (k = 0.0755 min$^{-1}$) was obtained at optimum conditions (pH = 11, O$_3$ flow rate = 0.019 mol h$^{-1}$, SPC concentration = 1 mol L$^{-1}$) during 60 min, while just 81% degradation effectiveness (k = 0.0323 min$^{-1}$) was achieved by H$_2$O$_2$–O$_3$ at the same conditions (H$_2$O$_2$ con-centration = 1.5 mol L$^{-1}$). The in-situ induced alkaline conditions and generated carbonate radicals as a result of the presence SPC led to a synergetic coefficient of 3.84 for SPC-O$_3$, proving the efficacy of this hybrid process. $^\bullet$OH, CO$_3^{\bullet-}$, O$_2^{\bullet-}$, and $^1$O$_2$ were the reactive species participating in degradation of BPS verified by scavenger tests. An increase in SPC till an optimum value enhanced the degradation of BPS, and further increase caused a reduction in the degradation effectiveness. Nevertheless, in investigated ranges, increasing O$_3$ flow rate and pH continuously increased the degradation effectiveness. This AOP system were revealed also to be very effective for high salinity conditions, expert of nitrates, which significantly inhibited the degradation of BPA. So high concentration of nitrates is however uncommon and, in such scenarios, biological denitrification should be preferred as first stage. Typically occurring

anions concentration range, applied as $\frac{Anion}{BPS}$ to BPS ratio of 100, except for $CO_3^{2-}$, had a negligible effect on the degradation. Advantageously, presence of $CO_3^{2-}$ increased the first-order degradation rate constant from 0.0632 min$^{-1}$ to 0.0985 min$^{-1}$. Developed AOP system proved its applicability for real case scenario wastewater treatment processes.

## Credit statement

Conceptualization: GB; Methodology: ZA, GB; Project administration: GB; Supervision: GB; Visualization: ZA; Data analysis: ZA, MPR, GB, Writing – original draft: ZA,MPR, GB; Writing – review & editing: XS, ZW, MPR, GB.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## Data availability

No data was used for the research described in the article.

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