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# Enhanced degradation and mineralization of sulfamethoxazole by integrating gamma radiation with Fenton-like processes

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ABSTRACT

Degradation of sulfamethoxazole (SMX) by ionizing radiation combined with goethite (α-FeOOH) catalyzed Fenton-like process were studied under various conditions. The results showed that SMX could be degraded completely at 2 kGy with or without the addition of goethite. This process followed pseudo first-order kinetics model. The presence of goethite promoted the mineralization of SMX, and the mineralization ratio increased with the increase of goethite dosage. The decomposition of SMX in goethite/ionizing radiation system exhibited high activity at pH ranging from about 3.0 to 11.0. Acidic condition was more preferable than alkaline condition for the degradation of SMX. The presence of humic acid inhibited SMX degradation, due to the competition of hydroxyl radical with SMX molecules. The addition of tert butyl alcohol (TBA) resulted in the decrease of SMX removal efficiency, indicating that hydroxyl radical played the main role in this system. This investigation demonstrated that goethite combined with ionizing radiation has potential for enhancing the mineralization of toxic organic pollutants in water and wastewater.

## 1. Introduction

Antibiotics has attracted increasing concern recently for their mass production and extensive application. A great quantity of antibiotic-containing wastewater was formed during their production and utilization, but antibiotics are difficult to be effectively removed by the conventional wastewater treatment processes, and subsequently transfer to the environment (Wang and Wang, 2016; Park et al., 2017). Antibiotics has been frequently detected in aqueous environment, including wastewater (Rivera-Jaimes et al., 2018; Wang et al., 2019), effluent (Thai et al., 2018), sludge (Huang et al., 2019) and surface water (Danner et al., 2019). Their occurrence and existence in aquatic environments can result in the generation of antibiotic resistant bacteria (ARB) and antibiotic resistant genes (ARGs), which can be transmitted to human and pose serious threat to human health (Kummerer, 2009). Among antibiotics, sulfamethoxazole (SMX) is a representative sulfonamide-type antibiotic which is used for preventing and curing bacterial infections diseases (Wang and Wang, 2018b). Because of its persistence and toxicity, it cannot be effectively destructed by traditional biological treatment methods (Wang and Wang, 2019). Therefore, the development of effective methods for antibiotic treatment are desired.

Advanced oxidation processes (AOPs) are efficient for the degradation of refractory organic pollutants in the water and wastewater (Wang and Wang, 2018a; Wang and Bai, 2017; Wang and Xu, 2012). During advanced oxidation processes, hydroxyl radicals (·OH) with high redox potential (up to 2.8V) are produced, which can oxide toxic organic pollutants to small molecules effectively (Wan and Wang, 2017; Tang and Wang, 2018b). Among various advanced oxidation processes, ionizing radiation is promising and potential technology (Wang and Chu, 2016; Yu et al., 2010; Hu and Wang, 2007; Wang and Wang, 2007). During ionizing radiation process, antibiotics can be decomposed by both direct and indirect way. As for direst way, high energy gamma ray or electron beam directly attack antibiotic molecules. As for indirect way, gamma ray or electron beam will radiate and excite water molecule and then generate various active radicals including hydroxyl radicals (·OH), hydroxyl free radical (·H), hydrated electrons ($e_{aq}^-$), and hydrogen peroxide ($H_2O_2$). The main reaction during water radiolysis process are described as equation (1). The numbers in brackets represent the number of molecules generated or consumed after receiving per 100 eV energy at a pH range of 6.0–8.5 (G-value). Ionizing radiation has been applied to decompose various antibiotics due to its high efficiency, no selectivity and no secondary pollution in degradation of organic compounds. Chu et al. (2018) reported that 0.27 mmol/L penicillin G could be completely degraded at the dose of 2.5 kGy. We studied the degradation of erythromycin A in

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antibiotic fermentation residues, and the removal efficiency reached 86% with 10 kGy ionizing radiation (Shen et al., 2018). Changotra et al. (2019) observed that 99% of ofloxacin was removed at 3 kGy. We found that the degradation efficiency of sulfamethazine was about 95% at 1 kGy (Liu and Wang, 2013). Sayed et al. (2016) demonstrated that norfloxacin degradation rate reached 91% at 0.87 kGy. However, the mineralization rate of ionizing radiation is limited. We observed that the erythromycin removal efficiency was 100%, while the TOC removal efficiency was only 26 at 10 kGy (Chu et al., 2019). Zhuan and Wang (2019) reported that the TOC removal rate of sulfamethoxazole was 12.4% with 1.5 kGy irradiation. Liu et al. (2014) observed that with 5 kGy irradiation, TOC removal rate of sulphadiazine was only 8.3%.

$$H_2O \rightarrow \cdot OH(2.7) + e_{aq}^{-}(2.6) + H\cdot(0.55) + H_2(0.45) + H_2O_2(0.71) + H_3O^{+}(2.6) \quad (1)$$

The Fenton oxidation process is widely used to degrade toxic organic pollutants (Liu et al., 2018; Tang and Wang, 2018a). However, the homogeneous Fenton process is limited due to the requirement of low pH and the production of iron sludge. Thus, the heterogeneous Fenton process was proposed to overcome these drawbacks (Mirzaei et al., 2017). Goethite (α-FeOOH) as an abundant and environmentally friendly iron mineral is applied to heterogeneous Fenton catalyst. It has been reported that goethite (α-FeOOH) exhibits the high Fenton activity in heterogeneous Fenton process, such as decomposition of paracetamol (Mameri et al., 2016), methyl orange (Wang et al., 2015), p-chloronitrobenzene (pCNB) (Li et al., 2015), Orange G (Wu et al., 2012) and 2,4,4' -trichlorobiphenyl (PCB28) (Lin et al., 2014). However, sulfamethoxazole degradation by ionizing radiation in the presence of goethite (as Fenton-like catalyst) was not reported.

In this study, the goethite was added in sulfamethoxazole solution as heterogeneous Fenton catalyst before ionizing radiation because iron is abundant in nature and widely applied in environmental remediation (Liu and Wang, 2019). The influence of goethite dosage, SMX concentrations and pH values on the degradation efficiency was evaluated. The SMX mineralization in the presence of goethite was also investigated. Moreover, the effect of humic acid as co-existing chemical species was observed. Finally, dose constant of hydroxyl radicals (·OH) with and without goethite was determined.

## 2. Materials and methods

### 2.1. Chemicals

The sulfamethoxazole (SMX, purity > 98%) was bought from Aladdin Industrial Corporation (China). The SMX stock solution (200 mg/L) was prepared using deionized water. Goethite (α-FeOOH, > 99%) was purchased from Shandong Maojia Environmental Protection Technology Co., Ltd (China). $H_2SO_4$, NaOH and humic acid were obtained from Beijing Chemical Reagent (Beijing, China). Tert butyl alcohol (TBA) was bought from Beijing Yili Fine Chemical Co., Ltd (China).

### 2.2. Experimental procedures

$^{60}$Co radiation source with the radioactivity of $3.0 \times 10^{14}$ Bq, located in the Institute of Nuclear and New Energy Technology (INET), Tsinghua University was used for gamma radiation process. The SMX solution samples were irradiated at dose of 0.2–2.0 kGy.

### 2.3. Analytical methods

The concentration of SMX was determined by HPLC (Agilent 1200 Series, USA). Total organic carbon (TOC) of the solution was analyzed using a Multi N/C 2100 TOC/TN analyzer (Analytik Jena AG Corporation, Germany).

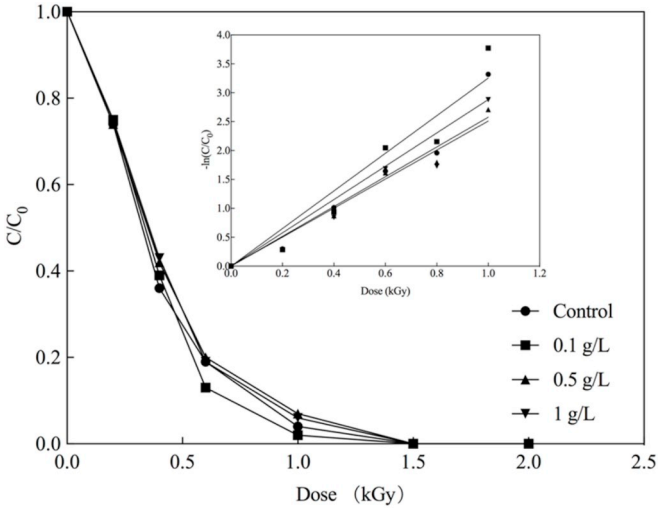


**Fig. 1.** Effects of α-FeOOH dosage on the degradation of SMX ($[SMX]_0$ = 20 mg/L).

## 3. Results and discussion

### 3.1. Degradation of SMX with the addition of α-FeOOH

SMX degradation by ionizing radiation is shown in Fig. 1. It can be seen that the degradation rate of SMX by ionizing radiation reached 100% after 1.5 kGy irradiation. The catalytic efficiency of α-FeOOH in the ionizing radiation process was also explored. The corresponding SMX removal efficiency also reached 100%with 1.5 kGy irradiation in the presence of various dosage α-FeOOH.

The first-order reaction rate constants were 3.059, 3.484, 2.619, and 2.748 $kGy^{-1}$ for the process when dosage of α-FeOOH was 0, 0.1, 0.5, 1 g/L (Fig. 1). The result indicated that the addition of 0.1 g/L α-FeOOH prompted the degradation of SMX compared with control group. During the degradation of SMX by gamma irradiation, a variety of active species including ·OH, ·H, $e_{aq}^{-}$ and $H_2O_2$ were generated by water radiolysis. The presence of α-FeOOH could prompt the decomposition of $H_2O_2$ to form ·OH as represented in equations (2)–(4), finally enhancing the degradation of SMX. The presence of ≡ Fe(III) will slowly react with $H_2O_2$ to generate ≡ Fe(II) by the following reactions as equations (2) and (3) (Kwan and Voelker, 2003). Then ≡ Fe(II) could react with $H_2O_2$ to form hydroxyl radical (·OH) as Fenton reaction. The hydroxyl radicals (·OH) could attack and destroy SMX molecules. Li et al. (2015) found that the addition of α-FeOOH could promote the degradation of p-chloronitrobenzene (pCNB). The removal rate increased from 10% by $H_2O_2$ oxidation alone to 80% by the heterogeneous Fenton-like process (α -FeOOH/$H_2O_2$). In the research by Wang et al. (2015), goethite catalyst showed higher catalytic activity compared with $H_2O_2$ alone and other catalysts. The decolourization efficiency of methyl orange improved from under 10%–98.9% within 70 min reaction in α -FeOOH/$H_2O_2$ Fenton-like system. Lin et al. (2014) investigated polychlorinated biphenyls (PCBs) degradation in a Fenton-like system using goethite as a catalyst. PCB28 was hardly removed by either $H_2O_2$ or goethite alone, whereas PCB28 removal rate reached about 99% in the presence of goethite and $H_2O_2$.

$$\equiv Fe(III) + H_2O_2 \rightarrow \equiv Fe(HO_2)^{2+} + H^{+} \quad (2)$$

$$\equiv Fe(HO_2)^{2+} \rightarrow \equiv Fe(II) + \cdot HO_2 \quad (3)$$

$$\equiv Fe(II) + H_2O_2 \rightarrow OH^{-} + \cdot OH + Fe(III) \quad (4)$$

The effect of α-FeOOH dosage on the kinetic rate of SMX degradation was shown in Fig. 1. When α-FeOOH dosage increased from 0.1 to 0.5 and 1 g/L, the reaction rate constants of SMX degradation decreased

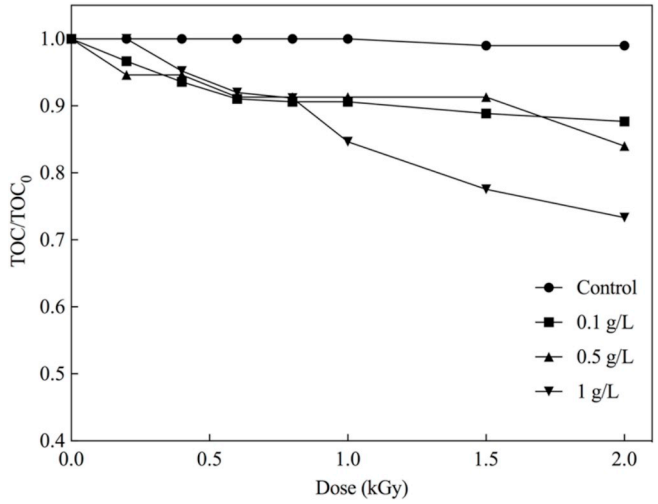


**Fig. 2.** Effects of α-FeOOH dosage on the mineralization of SMX ($[SMX]_0$ = 20 mg/L).

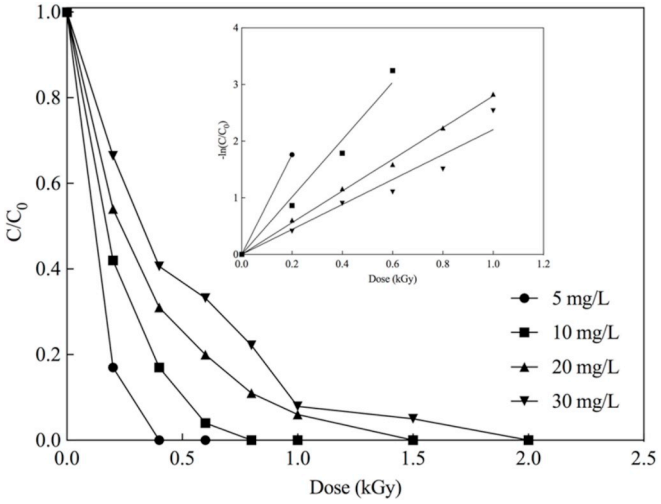


**Fig. 3.** Effects of SMX initial concentration on the degradation of SMX in the presence of α-FeOOH ([α-FeOOH] = 0.1 g/L).

from 3.484 to 2.619 and 2.748 $kGy^{-1}$, respectively. The increase of α-FeOOH catalyst dosage could supply more active sites for SMX decomposition, but result in the aggregation of catalyst particles (Ma et al., 2018). Moreover, the high goethite doses may result in the screen effect of goethite particles and lead to the decrease of degradation rate. In addition, the decrease of kinetic rate could be explained by the scavenger of hydroxyl radial by iron (Miller and Valentine, 1999; Huang et al., 2013).) The degradation rate of 2-chlorophenol (2-CP) decreased from 98% to 15% when goethite dosage varied from 0.5 g/L to 2 g/L during heterogeneous Fenton system (de la Plata et al., 2010). Huang et al. (2013) also found that bisphenol A oxidation rate increased when goethite dosage increased from 0.1 g/L to 0.25 g/L, then it decreased at higher goethite dosage during heterogeneous photo-Fenton oxidation process. Wu et al. (2012) reported the decolourization rate of Orange G declined when goethite dosage varied from 1.0 g/L to 1.2 g/L.

The mineralization of SMX (in terms of TOC removal efficiency) was also determined during the radiation-induced degradation of SMX. The addition of goethite prompted the mineralization of SMX by gamma radiation. As shown in Fig. 2, little TOC removal rate was observed for control group. By contrast, about 12.3%, 16.0% and 26.7% of TOC were removed after adding 0.1, 0.5 and 1 g/L α-FeOOH, respectively. The mineralization rate increased significantly increasing goethite dosage. In the researched by Liu et al. (2014), the addition of $Fe^{2+}$ enhanced the mineralization of sulfamethazine. This result may be due to $Fe^{2+}$ react with $H_2O_2$ generating hydroxyl radial. Sanchez-Sanchez et al. (2007) also found that higher goethite dose exhibited higher mineralization of aniline during electro-Fenton process. The more goethite addition, the higher $Fe^{2+}$ concentration in solution was determined. $Fe^{2+}$ could react with $H_2O_2$ formed by water radiolysis to produce hydroxyl radial and then decompose pollutants as well as intermediate products, finally resulting in higher mineralization rate.

### 3.2. Effects of SMX initial concentration

The initial concentration of SMX is an important parameter influencing the degradation of SMX by gamma irradiation. The initial SMX concentrations were set at 5, 10, 20, 30 mg/L, respectively, and the goethite addition was 0.1 g/L. Fig. 3 demonstrated the SMX degradation at various initial concentrations. The SMX degradation efficiency dropped with increasing initial SMX concentration. After 0.6 kGy irradiation, the removal efficiency reached 100%, 96.1%, 79.6% and 66.8% when the initial concentration was 5, 10, 20 and 30 mg/L, respectively. The degradation of SMX followed pseudo-first-order kinetics

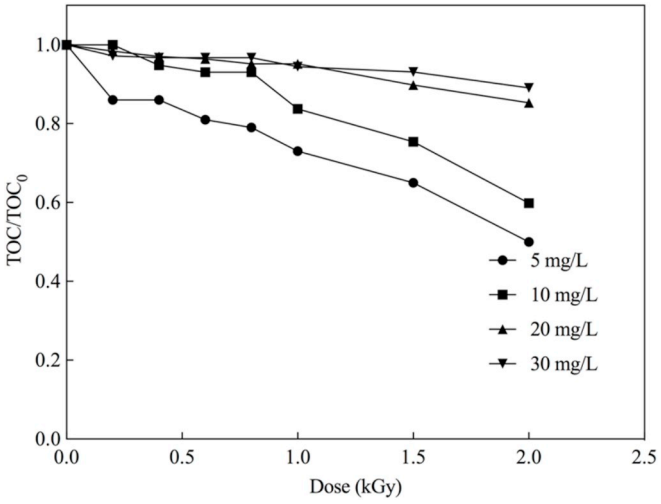


**Fig. 4.** Effects of SMX initial concentration on the mineralization of SMX in the presence of α-FeOOH ([α-FeOOH] = 0.1 g/L).

model, as presented in Fig. 3. The dose constant of SMX decreased from 8.82 to 5.06, 2.80 and 2.20 kGy -1 when the initial concentration increased from 5, 10, 20–30 mg/L, respectively. Fig. 4 showed the TOC removal efficiency at different SMX concentration with addition of 0.1 g/L goethite. It decreased from 50.1% to 40.2%, 14.7%, 11.0% when the initial concentration increased from 5 to 30 mg/L. The mineralization rate without the addition of goethite was 23.8%, 20.6%, 9.2% and 2.5% when the SMX initial concentration was 5, 10, 20–30 mg/L, respectively. The addition of goethite enhanced the mineralization of SMX. It was also found that the degradation of methyl orange declined with the increase of initial concentration (Wang et al., 2015). Hydroxyl radical (·OH) as one non-selective oxidant radical could oxidize SMX as well as its' intermediate products. With the increasing of SMX initial concentration, more intermediate products would compete with SMX molecule for ·OH, finally resulting in the decline of SMX decomposition rate (Wang et al., 2015).

### 3.3. Effects of pH

The pH value is an important parameter in Fenton process. Fig. 5 demonstrated the decomposition of SMX at initial pH from 3.06 to 10.98, respectively, when the SMX concentration was 20 mg/L and

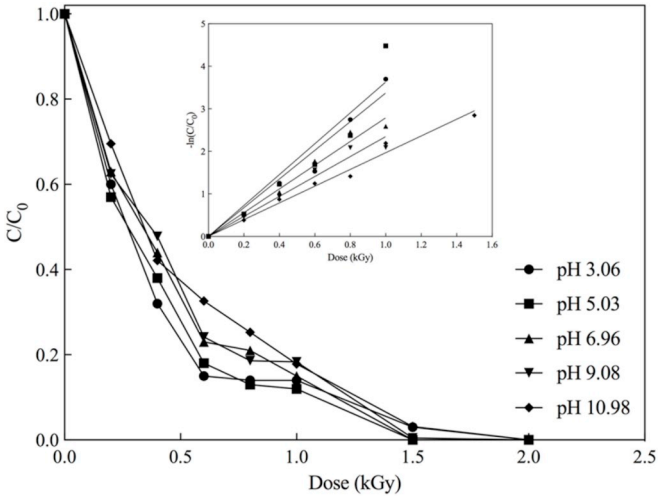


**Fig. 5.** Effects of pH on the degradation of SMX in the presence of α-FeOOH ($[SMX]_0$ = 20 mg/L, [α-FeOOH] = 0.1 g/L).

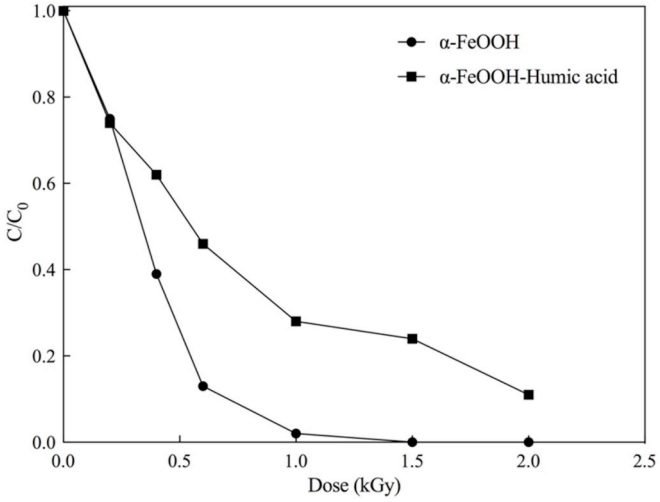


**Fig. 6.** Effects of humic acid on the degradation of SMX in the presence of α-FeOOH ($[SMX]_0$ = 20 mg/L, [α-FeOOH] = 0.1 g/L).

geothite dosage was 0.1 g/L. SMX could be degraded by gamma irradiation in a wide range of pH values. The decomposition efficiency all reached 100% after 2 kGy irradiation. As shown in Fig. 5, the dose constant of SMX degradation was 3.37, 3.63, 2.79, 2.35 and 1.97 $kGy^{-1}$ when pH increased from 3.06 to 5.03, 6.96, 9.08 and 10.98, respectively. The dose constant dropped as the increase of pH. The degradation of SMX with the addition of goethite exhibited better in acid condition. On the one hand, acid condition was more suitable for the form of soluble Fe from goethite (Huang et al., 2013). And then more hydroxyl radial would generate through the decomposition of $H_2O_2$ catalyzed by $Fe^{2+}$. The research by Lin et al. (2015) demonstrated that more hydroxyl radial was produced in acid condition during the goethite-catalyzed Fenton-like reaction. On the other hand, in alkaline condition, $OH^-$ could react with hydroxyl radial at a rate of $1.3 \times 10^{10}$L/(mol·s), decreasing the concentration of hydroxyl radial and inhibiting SMX degradation. It was also observed that the decolourization efficiency of methyl orange declined when initial pH was increased from 3 to 7 during goethite catalytic heterogeneous Fenton process (Wang et al., 2015). Lin et al. (2014) reported that 2,4,4' -trichlorobiphenyl (PCB28) removal rate declined from 99% to 52% as pH increased from 3 to 7 during the Fenton-like process catalyzed by α-FeOOH. In the study by Wu et al. (2012), higher decolourization efficiency of Orange G was observed at lower pH condition in goethite catalyzed heterogeneous Fenton-like reaction.

### 3.4. Effects of humic acid

The co-existing substance in solution may influence the degradation efficiency. The influence of humic acid on SMX degradation was investigated (Fig. 6). The presence of humic acid could inhibit SMX degradation by ionizing radiation. At the dose of 2 kGy, the SMX removal efficiency decreased from 100% to 89%. The presence of humic acid would compete hydroxyl radical (·OH) with SMX molecules, resulting in the drop of degradation rate. Guo et al. (2015) also observed that ciprofloxacin degradation efficiency decreased from 95% to 76.8% with 15 mg/L humic acid addition. Wang et al. (2017) reported that humic acid could scavenge hydroxyl radical and inhibited the degradation of amoxicillin, ofloxacin and cefradine by high energy electron beam irradiation. The effect of humic acid on the decomposition of p-

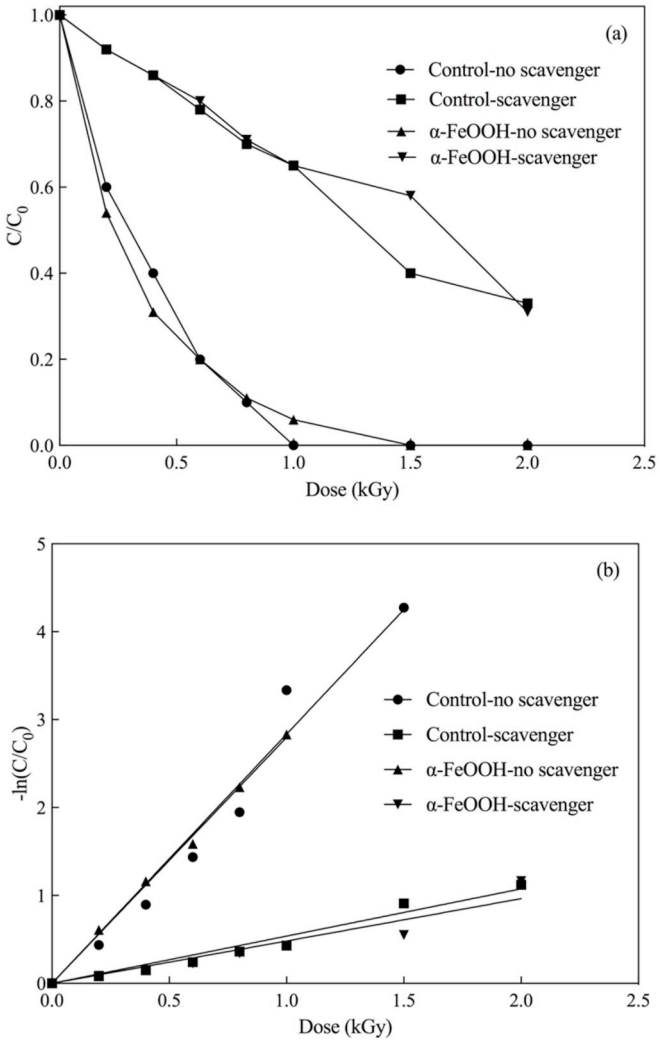


**Fig. 7.** Effects of tert-butyl alcohol on the degradation of SMX in the presence of α-FeOOH ($[SMX]_0$ = 20 mg/L, [α-FeOOH] = 0.1 g/L).

chloronitrobenzene (pCNB) by goethite catalytic heterogeneous Fenton-like process was studied by Li et al. (2015), and they found that various concentrations of humic acid all inhibited the removal of p-chloronitrobenzene (pCNB).

### 3.5. Role of ·OH radicals in sulfamethoxazole degradation

For exploring the role of OH radical played in SMX degradation, 100 mmol/L tert- butyl alcohol as radical scavenger was added in SMX solution before irradiation. The result was demonstrated in Fig. 7. Tert-butyl alcohol can react with hydroxyl radical (·OH) as following equation (Xu and Wang, 2012).

$$\cdot OH + \text{t-BuOH} \rightarrow \cdot CH_2C(CH_3)_2OH + H_2O \quad k = 6.0 \times 10^8 (L/(mol \cdot s)) \tag{5}$$

The corresponding dose constants were calculated by pseudo-first-order kinetic plots as following:

$k(\text{Control}) = 3.059\ kGy^{-1}$
$k(\text{Control-scavenger}) = 0.5388\ kGy^{-1}$
$k(\alpha\text{-FeOOH}) = 3.484\ kGy^{-1}$
$k(\alpha\text{-FeOOH -scavenger}) = 0.4819\ kGy^{-1}$
So, the dose constants for ·OH radical can be calculated as:
$k(\text{Control-}\cdot OH) = k(\text{Control})\text{-}k(\text{Control-scavenger}) = 3.059\text{–}0.5388 = 2.5202\ kGy^{-1}$
$k(\alpha\text{-FeOOH -}\cdot OH) = k(\alpha\text{-FeOOH})\text{-}k(\alpha\text{-FeOOH -scavenger}) = 3.484\text{–}0.4819 = 3.0021\ kGy^{-1}$

These calculation results revealed that when adding 0.1 g/L α-FeOOH, the dose constant of ·OH enhanced from $2.5202\ kGy^{-1}$–$3.0021\ kGy^{-1}$ compared control group. Thus, with the addition of α-FeOOH, more ·OH radicals were formed, resulting in higher SMX and TOC removal rate. Lin et al. (2015) found that ·OH was generated through $H_2O_2$ decomposition by the catalysis of α-FeOOH. Muruganandham et al. (2007) reported the addition of t-tubanol decreased the degradation rate of Direct Orange 39 in goethite catalyzed ultrasound irradiation system, indicating that hydroxyl radical played a main role in this process. Zhang et al. (2009) found the hydroxyl radical was the main active species for C.I. Acid Orange 7 degradation by ultrasound/goethite/$H_2O_2$ process.

## 4. Conclusion

Sulfamethoxazole could be effectively removed through ionizing radiation with the addition of goethite. The degradation efficiency of SMX reached 100% with various dosages of goethite addition. The presence of goethite enhanced the mineralization of SMX, TOC removal was 26.7% with 1 g/L goethite addition. The removal efficeincy of SMX with goethite exhibited high activity at a wide pH range from 3.0 to 11.0. The addition of free radical scavenger (tert-butyl alcohol) decreased the degradation of SMX, suggesting that hydroxyl radical was the dominant species in goethite catalyzed ionizing radiation process. The addition of goethite could enhance the reaction with $H_2O_2$ produced by water radiolysis and generate hydroxyl radical, finally increased the degradation and mineralization of SMX.

## Acknowledgements

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